

A Career in Catalysis: Alexis T. Bell

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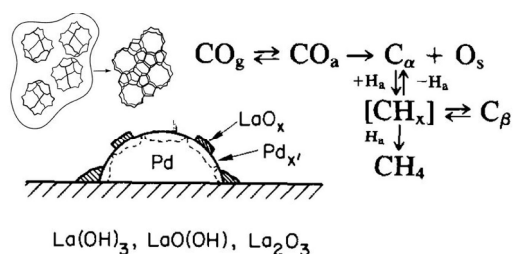
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Abstract

On the occasion of Alexis T. Bell's fiftieth year at Berkeley, we are honored to discuss a few aspects of his extensive contributions to catalysis, reaction engineering, and understanding of molecular scale structure in catalytic processes. The illustrations provided here help reveal some of his traits most valued by our community: a drive to employ the best methods of instrumental and computational analysis available; the instinct to search for the essence of the most important problems at hand, and the skill to write about them with exceptional clarity; and the formation and nurturing of collaborative teams to focus on the most essential questions.



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Introduction

The authors assembled for this account of course do not presume to strive for an overview of Alex Bell's contributions of 50 years. With around 700 papers and around 180 students and postdoctoral associates (not even trying to count how many active collaborators!),

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2
3 his career to date spans such broad and important problems that it would require a substantial
4 Festschrift to even attempt to do any justice to the work (Supporting Information). Nevertheless,
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6 as a collection of former group members spanning several decades (2010, 2004, 1998, 1987,
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8 1984, and 1979), rather than being comprehensive we aim to share a representative set of
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10 vignettes - a window into Alex' approaches and patterns that, we think, illustrate his enduring
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12 impact on the field. These are arranged roughly chronologically, addressing chemical vapor
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14 deposition, characterization of catalysts and catalytic processes, CO hydrogenation on supported
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16 metals, zeolite synthesis and design, partial oxidation on metal oxides, and quantum mechanical
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18 and statistical mechanical computation.
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25 With these vignettes, too, we hope to show glimpses of Alex's keen insight and
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27 mentoring expertise that stand out for us, certainly influencing our own careers and interests, but
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29 more broadly also establishing currents in the whole catalysis community. Finally, we briefly
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31 reflect on his personal influence on generations of students and collaborators. We offer
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33 apologies in advance to our many peers who could easily offer even more compelling and
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35 interesting stories about Alex's influence and contributions, but we are confident that they would
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37 all join us in celebrating his achievements and continuing to pass on the lessons learned from
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39 him.
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43 44 **The Early Years**

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46 Alexis T. Bell's earliest professional contributions were in the reaction engineering of
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48 plasma processes. His research over the first decade explored the mechanisms and kinetics of
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50 the plasma-enhanced chemical vapor deposition of inorganic and organic films. Through a series
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52 of papers published throughout the 70's and early 80's, Alex applied reaction engineering
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54 modeling to advance mechanistic descriptions of plasma chemistry and thin film growth (Figures
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1 and 2).^{e.g.,1-10} In this work, we see the emergence of Alex's approach towards scientific research: identify the important questions; master the spectroscopic and numerical tools needed to address the problem; determine the detailed reaction chemistry; and then share with the community the relationship between the reaction chemistry and process performance.

Table I
Reaction Mechanism for Plasma Polymerization of Unsaturated Hydrocarbons^a

Initiation	Adsorption	Termination
1. $e + M_g \rightarrow M'_g + H_2 + e$	7. $S + \begin{Bmatrix} M_g \\ M'_g \end{Bmatrix} \rightarrow \begin{Bmatrix} M_s \\ M'_s \end{Bmatrix}$	13. $R'_{gm} + H \cdot \rightarrow P_{gm}$
2. $e + M_g \rightarrow M'_g + 2H \cdot + e$	8. $S + H \cdot \rightarrow H_s$	14. $R'_{gm} + R'_{gn} \rightarrow P_{gm+gn}$
3. $e + M_g \rightarrow 2R'_{g} + e$	9. $S + R'_{gn} \rightarrow R'_{sn}$	15. $R'_{gm} + R'_{sn} \rightarrow P_{sm+sn}$
4. $e + H_2 \rightarrow 2H \cdot + e$		16. $R'_{sm} + R'_{sn} \rightarrow P_{sm+sn}$
Propagation (homogeneous)	Propagation (heterogeneous)	Reinitiation
5. $H \cdot + \begin{Bmatrix} M_g \\ M'_g \end{Bmatrix} \rightarrow R'_{g1}$	10. $R'_{sn} + \begin{Bmatrix} M_g \\ M'_g \end{Bmatrix} \rightarrow R'_{sn+1}$	17. $e + P_{gm+gn} \rightarrow R'_{gm} + R'_{gn}$
6. $R'_{gn} + \begin{Bmatrix} M_g \\ M'_g \end{Bmatrix} \rightarrow R'_{gn+1}$	11. $R'_{sn} + \begin{Bmatrix} M_s \\ M'_s \end{Bmatrix} \rightarrow R'_{sn+1}$	18. $P_s \xrightarrow{e, h\nu, I^+} R'_{sm} + R'_{sn}$
	12. $H_s + H \cdot \rightarrow H_2$	19. $H \cdot + P_{gn} \rightarrow R'_{gn} + H_2$
		20. $H \cdot + P_{sn} \rightarrow R'_{sn} + H_2$

^a M_g , primary monomer; R'_{g} , gas phase free radical; P_g , gas phase oligomer; M'_g , derivative monomer; R'_{s} , surface free radical; P_s , polymer fragment; $H \cdot$, hydrogen atom.

Figure 1. Sequence of elementary reactions expected to occur when a hydrocarbon monomer enters a discharge zone of a DC plasma reactor.⁸

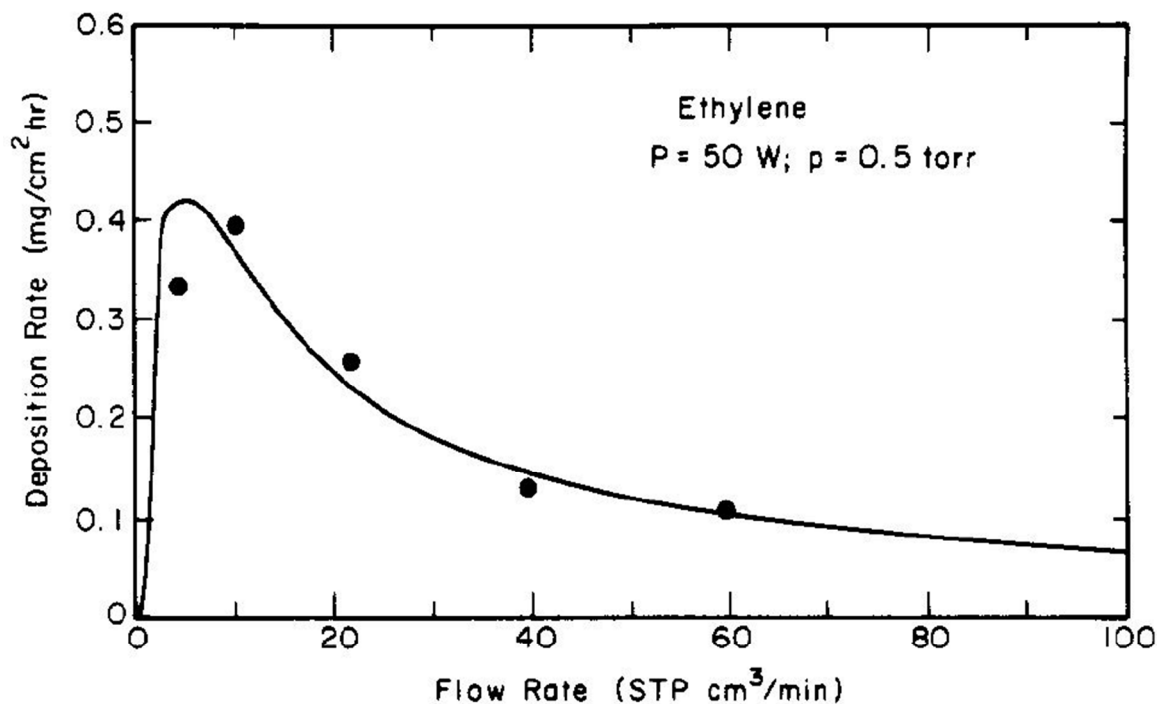


Figure 2. The rate of polymerization, which is the sum of gas and surface propagation steps was determined by solving the material balance equations that describe the monomer and free radical concentrations as a function of axial and radial position in the flow reactor. Predicted and experimental rates of polymer deposition as a function of monomer flow rate are one illustration of this model fit.⁸

Air pollution and energy security were major societal challenges of the early 70's. This created opportunities for exciting new research. Catalysis offered great promise in solving two major societal needs: (1) removing harmful pollutants from automotive exhaust; and (2) producing synthetic fuels to meet the energy crisis created by the U.S. oil embargo. Thus began the research program that is the foundation of Alex's career in catalysis. He started by addressing nitric oxide reduction^{11,12} and then moved on to Fischer-Tropsch synthesis and carbon monoxide hydrogenation to methanol.¹³⁻¹⁸

Alex's key motivation was to understand how catalyst structure affects selectivity, activity, and yield. Taking to heart the message over Latimer lecture hall, *i.e.*, that "progress in

science is based on advances in instrumentation”, he built a state-of-the-art laboratory for probing the active sites on catalysts. *In situ* infrared (IR) spectroscopy was employed during adsorption and reaction of NO and CO on supported precious metal catalysts (Figure 3). *e.g.*, 14,19-25 He developed the first micro-reactor for observing the infrared spectra of adsorbed NO and CO under the high-pressure conditions employed in Fischer-Tropsch synthesis.²⁶⁻²⁸ Dramatic changes in crystallite surface structure were discovered, depending on the composition of the metal oxide support and the method of preparing the catalyst. *In situ* Raman spectroscopy and NMR techniques were added to the laboratory as well.^{*e.g.*, 29-40} These techniques gave even greater insight into the evolution of the catalyst under reaction conditions. The Bell group discovered how metal crystallites transformed in the reaction environment, how they interacted with the support and additives, and how the nature of the active sites changed with time during catalysis.^{*e.g.*, 41-57}

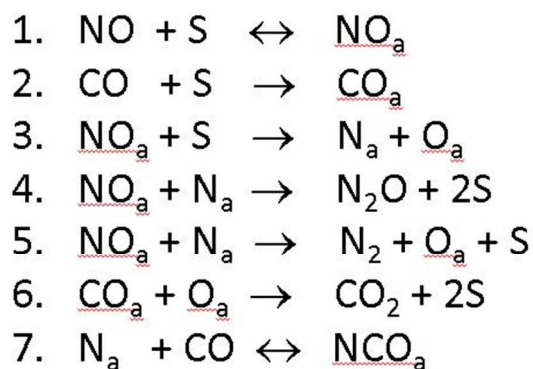
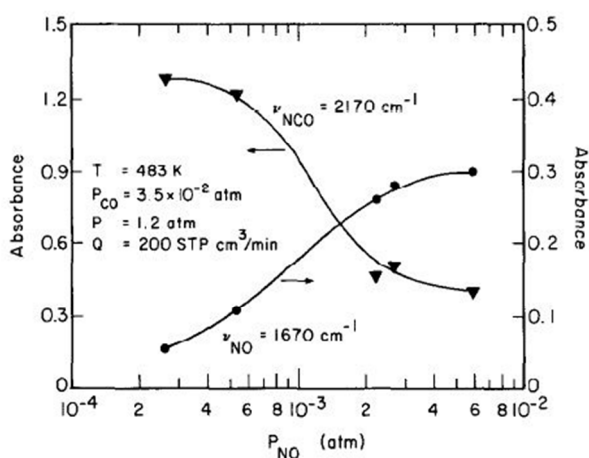


Figure 3. Dependence of the Rh-NCO and Rh-NO δ^- absorbance bands as a function of NO partial pressure. During the reduction of NO by CO over a Rh/SiO₂ catalyst an isocyanate group is formed on the surface of the rhodium microcrystallites. The dependence of the Rh-NCO band intensity on partial pressures of NO and CO can be interpreted with the aid of the reaction mechanism.²⁴

Alex pioneered transient techniques to complement his work on steady-state reaction rate chemistry. He and his students investigated the response of the adsorbates and the reaction products to temperature transients and to isotopic substitution. This allowed Alex to refine the reaction mechanisms proposed in his earlier work.^{e.g., 27,28,58-64} When coupled with *in situ* IR, Raman and NMR spectroscopy, he was able to elucidate the key rate-determining steps occurring on the surface of the metal catalysts, and to distinguish between the active and the spectator adsorbates (Figure 4).^{29,31,61,65} This combination of experimental techniques – developed and advanced by Alex, his students and collaborators – contributed to our current molecular-level understanding of relationship between catalyst structure and its function.

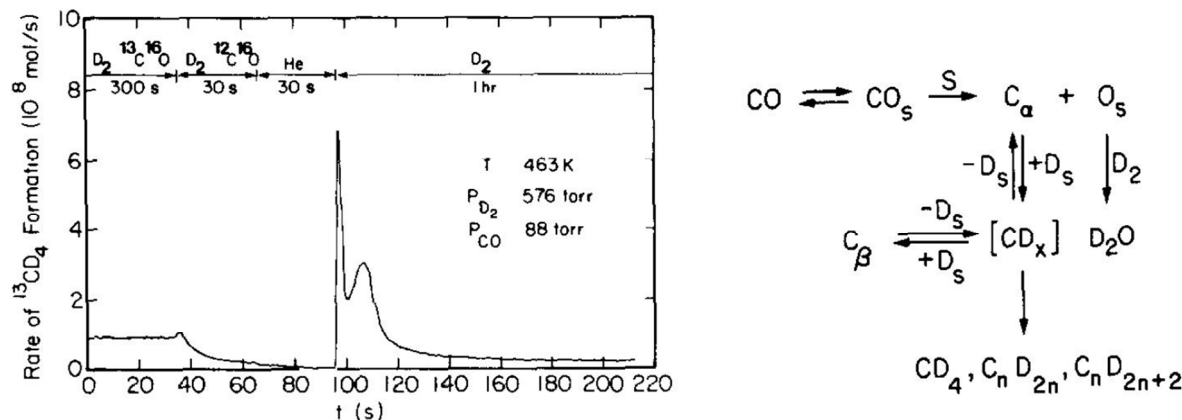


Figure 4. Transient response and isotope tracer studies revealed two distinctly different forms of carbon formed on Ru/SiO₂ during CO hydrogenation. C_α is highly reactive and is the principle precursor to C₂₊ hydrocarbons and methane. C_β accumulates slower than C_α and resides on Ru and the support.²⁷

A hallmark of Alex's research program was to bring together the best researchers, and share their ideas on how to tackle the many unanswered questions in catalysis. The Bell and Somorjai groups met weekly at Lawrence Berkeley National Laboratory to discuss their latest achievements. Structure sensitivity, intrinsic kinetics, and catalyst design were often the focus of these meetings. Several times a year, Alex Bell and Michel Boudart's students would meet to

1
2
3 compare the latest results from their respective labs. Students discussed the importance of
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5 identifying the active sites, looking for rate-limiting steps, and presenting rates in terms of
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7 turnover numbers. Detente between the USSR and the USA created a special opportunity to
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9 collaborate with scientists from the Institute of Catalysis in Novosibirsk. Doctors Davydov,
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11 Kuznetsov and Ryndin came to Berkeley and brought exciting new approaches to catalyst
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13 synthesis.⁶⁶⁻⁶⁹ [Yuri Ryndin even taught members of the group how to drink liquid nitrogen
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15 straight out of the Dewar!] Alex's students benefited here again from his mentorship. He taught
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17 us that scientific inquiry is a team effort; that to make breakthroughs one must embrace different
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19 viewpoints and learn new methods. Collaboration is key to staying at the forefront of
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21 engineering innovation and research.
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28 **Carbon Monoxide Hydrogenation on Supported Metal Catalysts**

29
30 One guiding principle in Alex's overall approach to understanding Fischer-Tropsch and
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32 methanol synthesis is that catalyst science is advanced through understanding the structure of
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34 adsorbed species and the dynamics of elementary processes involved in catalyzed reactions.
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36 Starting with a description of the reaction pathways he and his students worked to develop a
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38 detailed mechanism based on establishing the adsorbed species under reaction conditions and the
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40 chemical nature of the key reaction intermediate(s) in the process. This mechanism was then
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42 tested and refined against steady-state and transient measurements to establish the dynamics of
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44 the elementary processes. The mechanistic and dynamics insight also allowed the group to
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46 address the nature of the active site and how the catalyst could be designed to improve activity,
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48 selectivity, and in turn, yield of the desired products.
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54 Infrared observations,^{14,22} reactive scavenging^{15,70} and kinetic isotope effects^{16,59} led to a
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56 mechanism that has CO dissociating and carbon being hydrogenated to methylene and methyl
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groups, two key reaction intermediates (Fig 5). Hydrogenation of methyl produces methane and insertion of methylene into methyl produces an alkyl group that can continue to increase in carbon number by the insertion of additional methylene groups. During the early studies, it was noted that carbon was accumulating on the surface of the catalyst and not all the carbon was directly converted into products as indicated in Fig 5. This led to the proposal^{17,27,32,59,65,71} that carbon resided in a carbidic form, C_α , and an alkyl form, C_β (Fig 6).

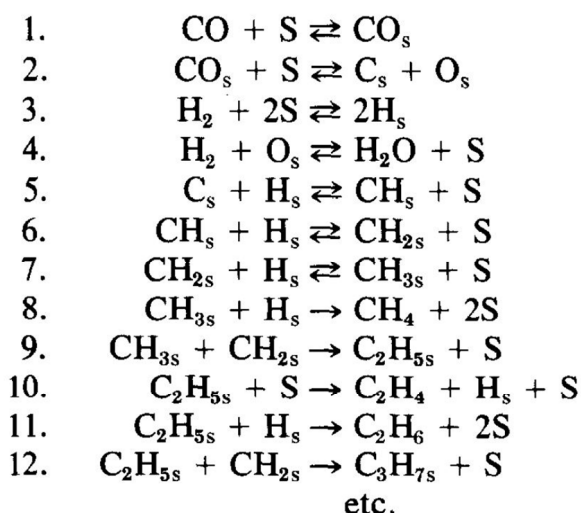


Figure 5. Proposed mechanism of hydrocarbon synthesis from CO and H_2 .¹⁸

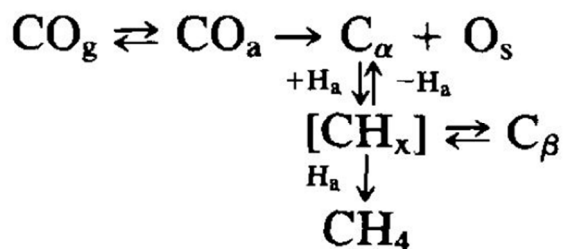


Figure 6. Reaction scheme for the rapid equilibration of CO into adsorbed CO and the interconversion of dissociated CO into the various forms of carbon.⁶¹

Alex adapted transient techniques (temperature programmed surface reaction (TPSR) and isotope switching) and established collaborations to bring *in situ* NMR spectroscopy to bear on

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2
3 identifying where the carbon resided, and how the different forms of carbon interconverted on an
4 active catalyst.^{31,32} The NMR signatures for carbidic and alkyl carbon were established as well
5
6 as the carbon that migrated from the metal to the support. Rates of interconversion of the
7
8 carbidic and alkyl carbon were established with transient techniques.⁶¹ This understanding was
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10 used in later studies to improve selectivity and yield.
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15 Bell and coworkers discovered that methanol synthesis over palladium is strongly
16 dependent on the composition of the support.^{43,44,69} Shown in Figure 7 is the effect of
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18 temperature and support type on the turnover frequency for carbon monoxide conversion into
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20 methanol.⁶⁹ The intrinsic rate varies by over two orders of magnitude with the lowest rate
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22 observed for palladium black and the highest for palladium dispersed over lanthanum oxide.
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25 Characterization of the Pd on La₂O₃ by X-ray photoemission spectroscopy and infrared
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27 spectroscopy of adsorbed CO revealed that a thin covering of rare earth oxide migrates onto the
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29 metal crystallites. A schematic of this unique catalyst structure is shown in Figure 8.⁴⁴ The
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31 migration of partially reduced LaO_x species onto the palladium is evidenced by a negative shift
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33 in the Pd 3d_{5/2} binding energy by as much as 0.7 eV below that for Pd dispersed on a non-
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35 interacting support (*e.g.*, SiO₂).⁴³ In addition, the LaO_x species suppress carbon monoxide
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37 adsorption on the metal crystallites, and weaken the Pd-CO sigma bond strength.⁴⁴ Surprisingly,
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39 the extent of the metal-support interaction, *i.e.*, the fractional coverage of rare earth oxide on the
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41 palladium increases with the metal particle size.
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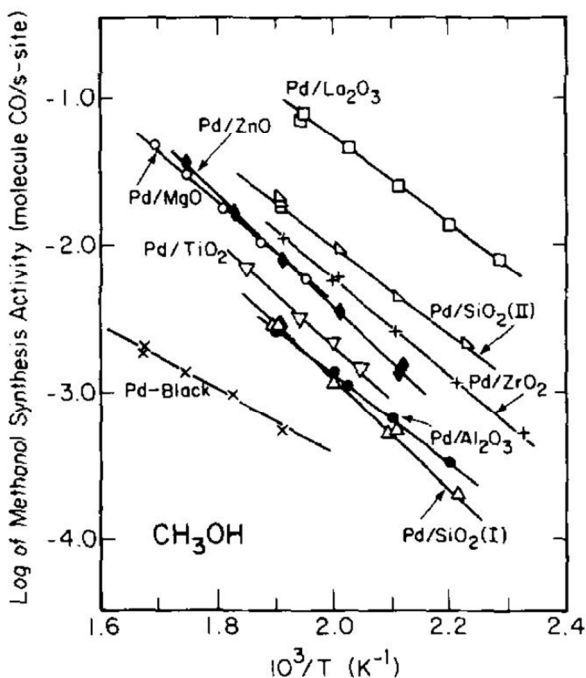


Figure 7. Arrhenius plots for the formation of methanol at 10 atm and a H_2/CO ratio of 3.⁶⁹

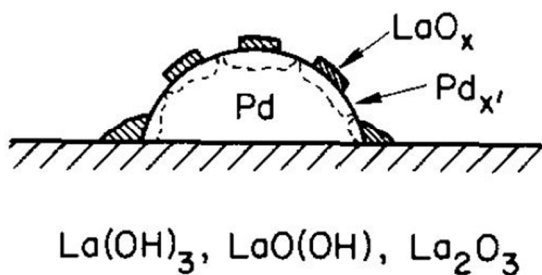


Figure 8. Schematic illustration of the interaction of the Pd with the support.⁴⁴

Bell's extensive work on carbon monoxide hydrogenation over supported metal catalysts reveals the true richness of catalytic reaction chemistry.⁷²⁻⁷⁵ It further illustrates the central role played by the catalyst structure on the reaction kinetics. The key to engineering better catalysts lies in the material and gaining molecular-level control over the synthesis of the active sites. This realization naturally led to Alex's pioneering work on zeolites.

Chemistry of Zeolite Synthesis

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4 Starting in the mid-1980's, Alex collaborated with Clay Radke, advising several students
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6 in projects to understand the hydrothermal synthesis of zeolites, with special attention to using
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8 the best tools available to gain insight into what governs the structure direction of zeolite and
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10 molecular sieve synthesis. The emphasis was on understanding the process at the molecular
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12 level, with an eye to learning how to direct and engineer catalyst sites with molecular control.
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16 In one theme, they pursued an understanding of the solution chemistry, especially using
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18 high resolution quantitative multinuclear liquid nuclear magnetic resonance spectroscopy to
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20 characterize the solution chemistry at play in synthesis of aluminosilicate zeolites such as zeolite
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22 Y, of templated highly siliceous zeolites such as ZSM-5 and silicalite, and of molecular sieves of
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24 more varied composition. *e.g.*, 33,76-90
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28 In another theme, they also used solid state magic angle spinning (MAS) NMR to gain
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30 understanding of the gel and nucleation processes. One series of papers addressed the role of
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32 seed gels in governing the structure of template-free aluminosilicate zeolite synthesis, such as is
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34 typical for zeolite Y.⁹¹⁻⁹³ In another series of papers, they pursued more particularly an
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36 understanding of the role of a template cation in directing the crystallization of silica-rich
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38 zeolites.^{87,94-97}
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42 Characteristically, Alex punctuated with a few periodic papers that give an update on the
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44 current thinking, reviewing how patterns and insights can be perceived from contributions from
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46 both his own and others groups to frame the next set of broad questions. We can take the
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48 opportunity with the series on silicalite to obtain a glimpse of how Alex approached this field
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50 and left a lasting impact. In the following, short excerpts from the papers illustrate how the
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52 problem – how zeolite nucleation and structure control is achieved - was clearly framed early on,
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3 attacked methodically, and after rigorous investigations brought to a new level of understanding
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5 and design challenges.
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8 Starting in 1991, with Clarence Chang⁹⁸ (then visiting professor from Mobil Research
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10 and Development, and a leading figure in templated-zeolite synthesis) Alex examined the then
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12 extent data, and new data, of ZSM-5 synthesis carefully to postulate more clearly than anyone
13
14 how templates might work. They stated that “embryonic structures are formed rapidly with
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16 heating”, which may have the stoichiometry associated with a template cation surrounded by the
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18 intersection structure (Figure 9) – about 1 tetrapropylammonium ion (TPA) to 24 silicons – in a
19
20 clathrate structure. These structures could be
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22

23
24 “randomly lined together, but in time become ordered or “annealed” through repeated
25
26 cleavage and recombination of the siloxane bonds, mediated by the OH⁻ ion” (Figure 9).
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30 This clear, detailed, and testable hypothesis added structure to the discussion in the
31
32 literature at the time, postulated by early leaders in the field such as Breck and Flanigen (nicely
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34 reviewed by Flanigen⁹⁹); the basis of the hypothesis is that “the isomorphism between clathrate
35
36 hydrates and certain framework silicates is exemplary”. Further, though, Alex and Clarence
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38 drew this implication – that the richness and variety of templated zeolite structures might be
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40 explained and even predicted, since the
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44 “structure of the solvation sphere around the organic template will be influenced by
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46 factors other than geometry, such as pH, temperature, ionic strength, inorganic ions, etc.
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48 so that the same template may give rise to different zeolites phases under different
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50 conditions”.
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54 This laid out the challenge of that decade, then, in this field – to better understand and predict the
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56 action of the template.
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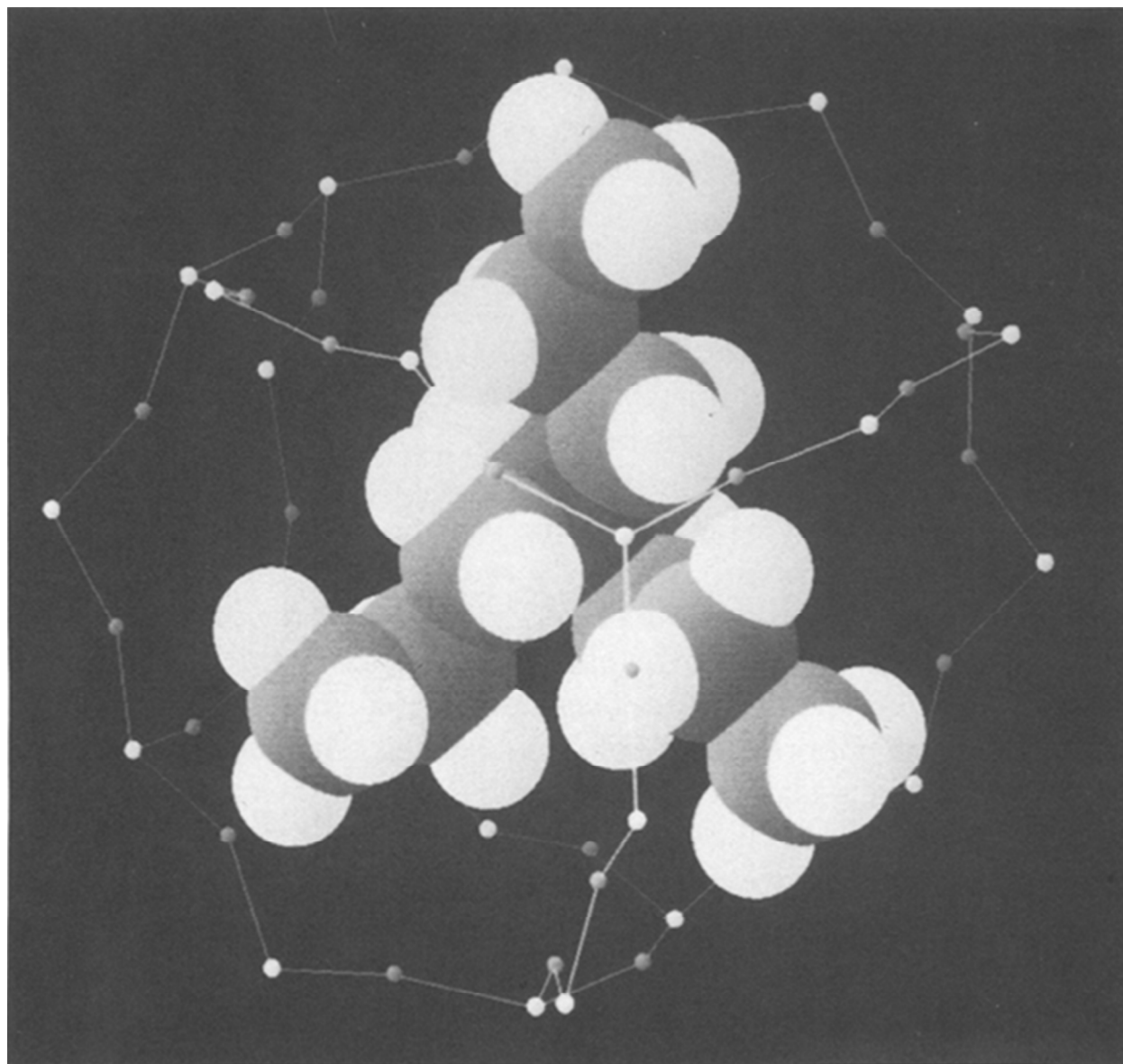


Figure 9. Illustration of clathrate organization around a template cation.⁹⁸

Alex pursued this challenge, seeking a rigorous understanding that could yield quantitative insights into structure direction. Silicalite (the purely siliceous isomorph of ZSM5) provided an excellent model and a rich set of contributions. By 1994, Alex was able to update the current thinking in another contribution, stating “the question is how do organic cations order the silica in zeolite synthesis gels form nuclei for zeolite crystallization”. He summarized the findings then current:

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3 “²⁹Si MAS NMR and ion-exchange experiments of TPA-containing sodium silicate
4
5 synthesis gels suggest that TPA is encapsulated in the gel in silicate cages prior to the
6
7 appearance of any crystalline material. Crystallization occurs within the gel via
8
9 rearrangement of the TPA-occluded silicate cages by the breaking and reformation of
10
11 siloxane bonds into the more stable silicalite structure”.

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14 Alex goes on to show that this understanding is consistent with the then-contemporary
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16 contributions from other groups using ¹³C NMR, ¹H-²⁹Si and ¹H-¹³C CP-MAS NMR, small angle
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18 neutron scattering (SANS), and Raman spectroscopy. Further, though, he again draws
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20 implications in the most precise terms of the time:
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24 “The evidence also suggests that to form a given zeolite, e.g., ZSM-5, the TAA cation
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26 should be as hydrophobic as possible, but not so large as to be unable to fit into the
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28 zeolite pore structure in an ordered array.”
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32 Even further, he suggests that these questions can and should be pursued with computation. He
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34 also later contributed to this computational pursuit as well.^{96,100,101}
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36
37 While encouraging the community along these lines, Alex and Clay made further
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39 contributions of their own through the 90’s, and Alex took the opportunity once again to give an
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41 update on current thinking and prospectus in 1999¹⁰² with a much more specific, quantitative,
42
43 experimentally tested, and molecularly detailed explanation of all the extent data, suggesting
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46 “the following scenario for the nucleation and growth of silicalite. The initial synthesis
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48 gel contains a highly articulated silicate network exhibiting a relatively low
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50 connectivity.... Hydrated TPA cations are trapped within pockets of the gel. Upon
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52 heating, the silicate network condenses.... This accounts for the decrease in the {tri-
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54 connected to tetra-connected site} ratio and the increase in pH of the mother liquor
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3 during the induction period. As the gel condenses, it becomes more hydrophobic and
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5 expels the water pockets along with the dissolved cations, accounting for the rise in the
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7 Si/TPA ratio in the solids. As the gel condenses further and its connectivity increases
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9 (Figure 10), the hydrophobic TPA cations are engulfed by the silica network, thus
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11 forming a cage around the TPA cation. In basic solutions, silica has a net negative
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13 charge, thus attracting the TPA cations by Coulombic forces. Occlusion of TPA in the
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15 silicate cages competes with the expulsion of solvated TPA contained within the water
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17 pockets in the gel. At the onset of crystallization, TPA occlusion dominates over the
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19 expulsion of solvated TPA, and as a consequence the Si/TPA ratio of the solid phase of
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21 the gel decreases.”
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27 – ultimately reaching the stoichiometric value explained with Clarence Chang back in 1991.
28

29 Figure 11 shows one of the key experiments – quantitative ^{29}Si MAS NMR demonstrating a
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31 distinct shift in connectivity of the network that marks the induction period. Reflecting on this
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33 excerpt a bit, we see Alex’s characteristic precision in cleaning up what had been a complicated
34
35 and murky process description, accounting carefully for the implications from his own work as
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37 well as others’ work. Each point in this carefully constructed explanation is backed up by data –
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39 much of it collected by Alex, Clay, and students following Alex’ formulation of the challenges in
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41 1991 and 1994.
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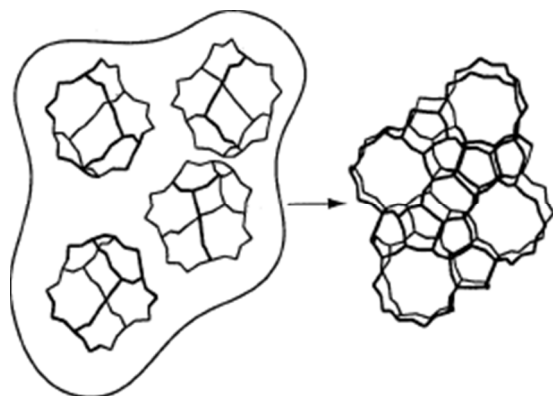


Figure 10. Assembly of clathrates into a prospective nucleus.⁹⁸

In that same review, Alex also was able to show patterns that clarified the current thinking of how aluminosilicates interact with templates to nucleate aluminum containing zeolites; the recent finding from both MAS and liquid NMR showed that the “aluminate anions appear to react with small silicate anions to produce aluminosilicate anions and cause a depolymerization of high molecular weight silicate structures”. This crucial insight helped explain how Al containing systems could manage to participate in the templating process described with the purely siliceous system. Alex and Clay also went on to further important contributions to address the solution behavior of an even richer variety of inorganic ion precursors and templates.

This vignette shows again how Alex worked in collaboration to formulate and attack key problems in a field of importance (here, zeolite synthesis work), driving toward advancing the current state of understanding using both his own and others’ insights, and to periodically provide a clear exposition of the next challenging set of questions for the whole community to address.

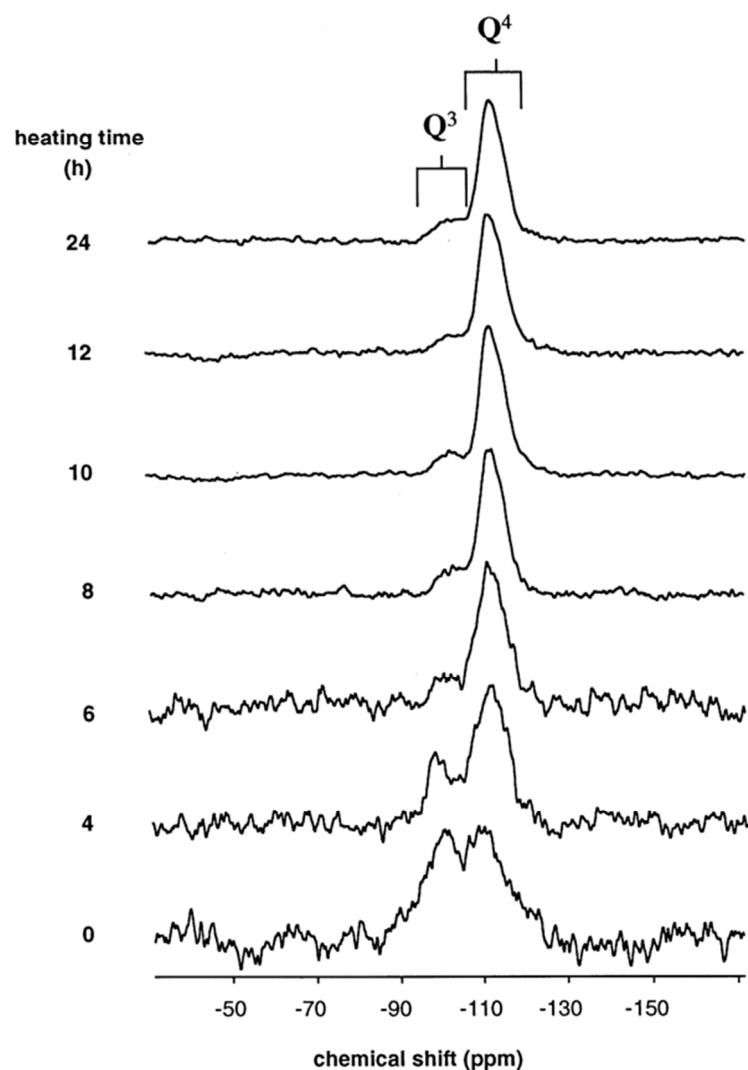


Figure 11. Quantitative ^{29}Si MAS NMR patterns, offering key evidence pointing toward a comprehensive description of the silicalite synthesis induction period.¹⁰²

Oxidation Catalysis on Metal Oxides

One is hard pressed to find either a catalytic moiety or a catalytic reaction that Alex has not touched. Still, certain thematic trends reappear more often than others. One particular family of catalytic materials that have yielded productive investigation spanning four decades has been supported vanadates,^{39,40,103-129} including collaboration with Enrique Iglesia to investigate the kinetics and mechanism for oxidative dehydrogenation of propane, and collaboration with T. Don Tilley to develop techniques for better control of deposition and anchoring of isolated

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2
3 vanadates on metal oxide surfaces. The ability to generate catalytically active sites with
4 structural uniformity using isolated vanadates that developed from these studies allowed Alex to
5 expand his effort in the area of “single-site heterogeneous catalysis” as applied to the oxidation
6 of methanol to formaldehyde.¹³⁰⁻¹⁴⁰
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12 Alex writes “A central objective in the field of catalysis is to establish relationships
13 between the composition and structure of catalytically active centers and their activity and
14 selectivity.¹³⁰ For most heterogeneous catalysts, this is a difficult task because of the
15 heterogeneity of active sites, making it nearly impossible to determine the environment of an
16 active site uniquely.” By exploiting the ability to anchor isolated VO₄ units onto the surfaces of
17 several metal oxide supports, Alex was able to pursue this objective of uniquely tying structure
18 and activity in a series of papers from the mid-2000s to the early 2010s that demonstrates his
19 careful and meticulously thorough approach to mechanistic work. By this time, Alex was able to
20 unleash the full capabilities of his now-mature techniques in *in situ* FTIR, Raman, and X-ray
21 absorption near edge spectroscopy (XANES) along with density functional theory (DFT)
22 calculations to leave no stone unturned in the pursuit of the definitive explanation that ties
23 together disparate data collected by many researchers working in the same area. The resulting
24 collection of papers reads a little like a multiperspective novel, with the reader’s attention
25 shifting back and forth between the point-of-view of two narrators, an experimental one and a
26 computational one. And, as with the fiction, the two narrators tell the same story in an
27 overlapping and complementary way.
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50 The story begins on the computational side with an exploration of how VO₄ interacts with
51 the metal atoms of the support.¹⁴¹ Isolated vanadates consist of a central vanadium atom with a
52 vanadyl oxygen (V=O) and three oxygens bridging with surface atoms (V-O-M). In this paper,
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3 the computational model included essentially only this basic structure (OH groups were added to
4 complete the valences of the M atoms), and varied the support (M = Si, Ti, Zr) to investigate the
5 support effect. It was already established in the experimental literature that the activity of
6 supported vanadates decreased in the order $\text{ZrO}_2 > \text{TiO}_2 > \text{SiO}_2$, though the reason for this order
7 was not established. While these early calculations did not reproduce the experimental results
8 quantitatively, they also failed to show any significant difference in either the apparent activation
9 energy or the pre-exponential factor for methanol oxidation over the different supports. This
10 paper concluded that the electronic properties of the metal in the support could not explain the
11 effects seen in the experiments, as they would have surely been apparent in the calculations. The
12 results also suggested that methanol adsorption formed V-O-CH₃ and M-O-H groups as they
13 were predicted to be much more stable than the reverse of M-O-CH₃ and V-O-H, but again
14 negated any contribution of M in differentiating between supports.
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32 These results created a puzzle – why did the supports show activity differences in
33 experiments but were nearly indistinguishable computationally? Alex would spend eight years
34 building a systematic case to explain these observations, but the original conclusions of his first
35 and simplest exploration of the subject were never refuted by his later work.
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42 Next, Alex set out to establish the definitive mechanism for methanol oxidation over
43 vanadate on silica (MCM-48).¹³⁰ Through a series of carefully planned *in situ* temperature
44 programmed spectroscopic experiments, the population of adsorbed and surface species were
45 tied to the gaseous products. Every bit of data in the paper supports the final conclusion, and the
46 paper is a great example of Alex's 'leave no stone unturned' approach. Every possible surface
47 interaction is tested and every competing theory is discussed. In the end, the totality of the FTIR,
48 Raman, XANES and kinetic measurements leads to a single mechanism. Importantly, this paper
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provides direct observation that proton transfer from methoxide to the vanadyl oxygen is the rate-limiting step – as suggested by earlier computational work. At the close of this experimental chapter, the evidence is summarized in two mechanistic schemes for the temperature programmed desorption (TPD) and temperature programmed oxidation (TPO) experiments that show the flux of each species and proposed intermediate (Figures 12 and 13). But these experiments were only able to observe the oxidation of methanol, which necessarily must have resulted in reduced vanadium centers that were not observed. Alex concluded that the reoxidation must have been fast, but no mechanism was known.

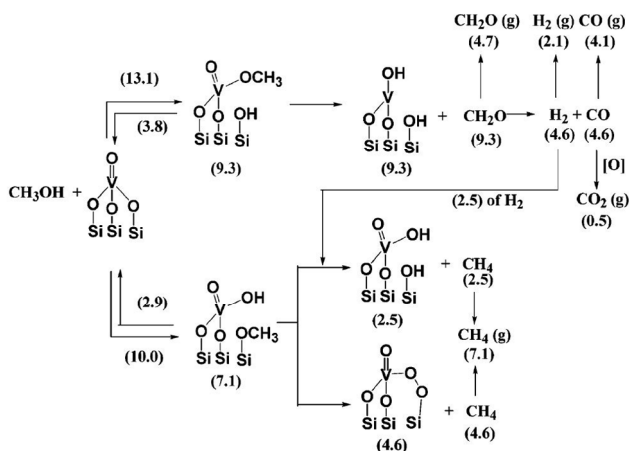


Figure 12. Mechanistic scheme for the TPD experiment on 3.4V/MCM-48.¹³⁰

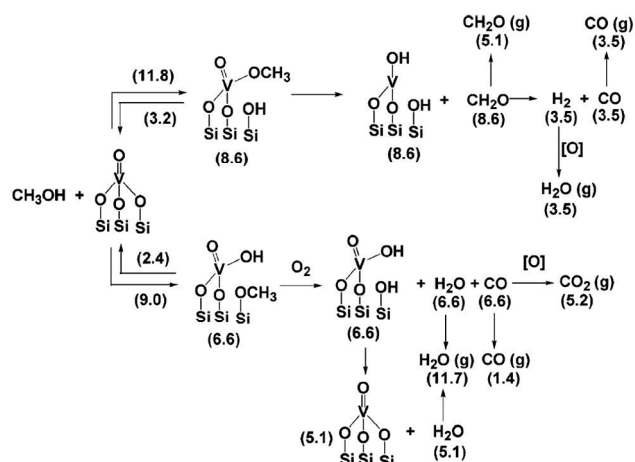


Figure 13. Mechanistic scheme for the TPO experiment on 3.4V/MCM-48.¹³⁰

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5 Naturally, the next paper in the series was about the reoxidation of the vanadium. If a
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7 process is too fast to observe experimentally, it is best approached computationally. An update to
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9 the support model for SiO₂ involved using a silsesquioxane molecule, with one Si-atom vertex
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11 replaced by V=O (Figure 14).¹³¹ With DFT, Alex was able to probe every step in the methanol
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13 oxidation/vandadium reduction mechanism and show that it matched the experimental results,
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15 but also add the missing details about the vanadium reoxidation mechanism. Once again, Alex's
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17 intuition proved correct, and the reoxidation of V³⁺ to V⁵⁺ was facile as compared to the
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19 methanol oxidation step, explaining why V³⁺ species were never observed. In the process, the
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21 paper touches upon every possible mechanism along the way, so as to eliminate the infeasible
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23 ones with evidence. Here also the reoxidation mechanism was established. The issue is that each
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25 molecule of dioxygen should oxidize two reduced vanadium centers, but the vanadium atoms are
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27 isolated from one another. The solution is to have molecular oxygen adsorb to the V³⁺, creating a
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29 peroxy group that will propagate an oxygen atom through the support Si-O-Si bonds until it
30
31 reaches another reduced V³⁺ center. The notion that oxygen atoms could travel through silica
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33 was counterintuitive, but was supported by both earlier work over isolated molybdates on
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35 silica¹⁴² and, of course, by Alex's own follow up paper,¹³³ where he showed the same isotopic
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37 scrambling using ¹⁸O₂ over isolated vanadates on silica.
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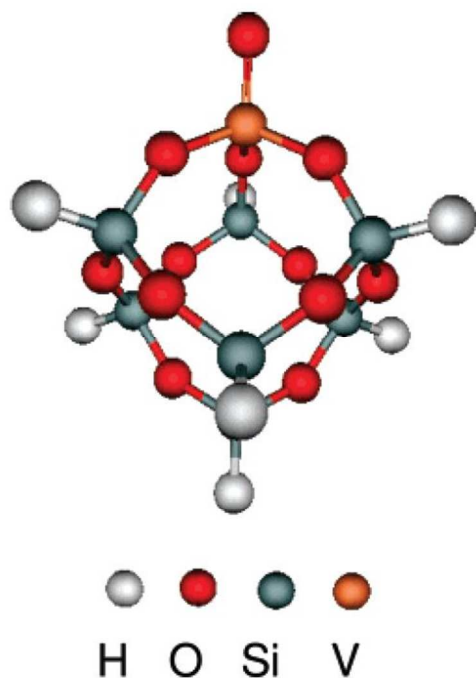


Figure 14. Model of the VO_x/SiO_2 catalytic site.¹³¹

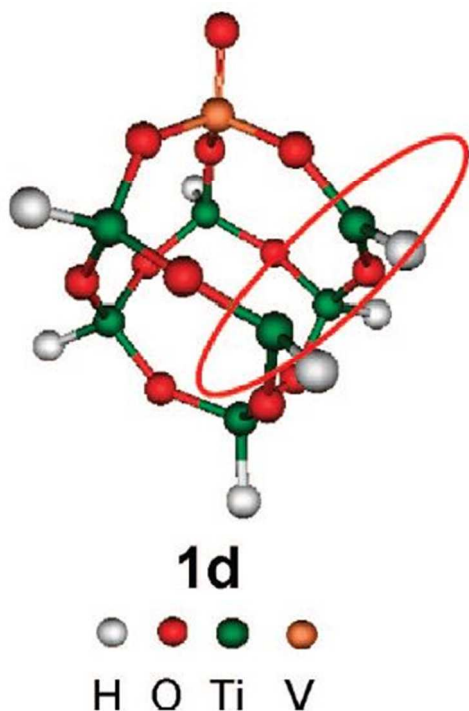


Figure 15. Model of the VO_x/TiO_2 catalytic site. The location of the O-vacancy is circled.¹³⁵

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3 Common threads found throughout Alex's work and exemplified in these papers are his
4 ability to anticipate questions and criticisms and provide responses in the same papers, and his
5 ability to predict correctly the findings of future papers in the same series.
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10 Having conquered the mechanism of methanol oxidation on VO_4/SiO_2 , the next logical
11 step was to understand why SiO_2 was such a poor support and why TiO_2 ¹³² and ZrO_2 ¹³⁶ were
12 three orders of magnitude higher in activity under the same conditions. The reactivity of these
13 supports is very different from silica, but how exactly does that make the methanol oxidation
14 reaction faster? When all three supports were treated as structurally the same¹⁴¹ calculations
15 suggested that the apparent activation energies for methanol oxidation would all fall between 112
16 and 123 kJ mol^{-1} . But the experiments showed that only SiO_2 (96 kJ mol^{-1}) was even close, with
17 TiO_2 (67 kJ mol^{-1}) and ZrO_2 (75 kJ mol^{-1}) quite a bit lower.
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29 First, the experimental work ruled out all other possible explanations, and together with
30 the computations, suggested that these surfaces are not structurally interchangeable. Of course,
31 Alex had a hypothesis, which he then followed up with a new computational study that explored
32 the role of oxygen vacancy defects on the activity for methane oxidation (Figure 15).¹³⁵ As it
33 turns out, the oxygen vacancies typically encountered on TiO_2 surfaces, when located adjacent to
34 a V-O-Ti bond, "provides the active site with more flexibility thereby allowing for a larger
35 degree of H-bonding in the [V-O- CH_3 dehydrogenation] product between Ti-OH and V-OH
36 ligands." The oxidation state and environment of vanadium are not changed, hence no
37 differences in the XANES or Raman spectra between SiO_2 and TiO_2 are observed. The surface
38 species involved are the same, hence no change in the FTIR spectra either. Such a puzzle as
39 support effects between SiO_2 and TiO_2 came down to a small difference in the flexibility of the
40 bonding in the support backbone, leading to stronger hydrogen bonding. This small difference
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3 had a big impact on the results. The stabilization of the transition state in the rate-limiting step
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5 lowered the calculated apparent activation energy to 66.5 kJ mol^{-1} , in excellent agreement with
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7 the experiment. To engineer this observation into a new catalyst, Alex later followed up this
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9 work with computational and experimental investigations of bilayered $\text{VO}_x/\text{MO}_x/\text{SiO}_2$ materials
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11 with defect forming metal oxides ($\text{M} = \text{Ti}, \text{Zr}, \text{Ce}$).¹³⁷⁻¹³⁹
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15 The story here developed from the ability to characterize the mechanism at single site
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17 catalysts, and under Alex's watch revealed how structural flexibility in defective metal oxides
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19 enhances transition state stabilization, and how new classes of catalysts could be developed to
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21 exploit these properties.
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25 **Alex in Theory**

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27 From the end of the 1980s on, Bell's work gradually includes more computational,
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29 quantum and statistical mechanical simulations to complement experimental work. As computers
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31 became more powerful, this was not a digression, but a natural expansion to achieve his overall
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33 objective to understand those fundamental underpinnings of catalysis that could be translated to
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35 the context of practical applications, for the purpose of catalyst development and design.
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39 His views are nicely outlined in a short perspective article in 1990⁷⁵ on the "impact of
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41 catalyst science on catalyst design and development," which is still relevant. Paraphrasing Bell,
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43 this impact results from: (1) the pursuit of scientific understanding, which continues to lead to
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45 the development of new analytical techniques, helping to reduce the time and options to evaluate
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47 catalysts by testing; (2) molecular level views (one could now add "nano"-level views, as in his
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49 perspective in *Science* in 2003),¹⁴³ to guide catalyst formulations; (3) development of new
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51 theories and concepts, to achieve relationships between catalyst structure and performance. This
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53 scientific approach reduces empiricism in development, and, increasingly, design.
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3 The chemical engineer in Bell is once again clear from his choice of reactions that are of
4 particular, timely industrial significance, in particular in the energy and environmental arena,
5 such as removal of NO, methanol or Fisher-Tropsch synthesis. Here, he attempts to obtain
6 information under conditions that are relevant to industrial catalysis. For the same reasons, *in situ*
7 spectroscopy is used to unravel reaction mechanisms and understand the structural and
8 compositional changes that occur in heterogeneous catalysts, in mixtures, under reaction
9 conditions. There is a preference for *in situ* IR and Raman, but also NMR and isotopic tracer
10 studies, to probe species over time, on different surfaces that aim to best represent the industrial
11 catalyst, or a model that would help to unravel pertinent principles.
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24 Alex Bell avidly collaborates with other chemical engineers and chemists at UC Berkeley
25 and LBNL, as well as internationally, to build on complementary expertise. These key
26 collaborations signal new directions, or help to obtain the missing piece in the catalytic puzzle.
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31 Alex' theoretical work began in the late 1980s with Evgeny Shustorovich from Eastman
32 Kodak in Rochester. They used the bond-order-conservation-Morse-potential method (BOC –
33 MP)^{74,144-151} to account for molecule-surface and intermolecular interactions at realistic loadings,
34 in order to study adsorption, surface diffusion and desorption by, for example, Monte-Carlo
35 techniques. In conjunction with the papers with Lombardo over the same period,^{64,103,152-154}
36 theoretical models appeared that would help to describe kinetics of gases on metal and other
37 surfaces. Importantly, results from these models could be tested against TPD and other
38 experimental methods. The 1991 review by Lombardo and Bell in *Surf. Sci. Rep.*¹⁵⁴ signaled a
39 new era in which various types of Monte-Carlo simulations, molecular dynamics, transition-state
40 theory, and other statistical physical as well as quantum mechanical approaches would
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increasingly play a central role in studying diffusion, adsorption and reaction in catalysts, in particular zeolites.

The second stage of Alex' computational work, a notable collaboration with Doros Theodorou, lasted throughout the 1990s. They developed powerful Monte Carlo tools for understanding adsorption and diffusion of molecules in zeolites.¹⁵⁵⁻¹⁵⁷ Alex and Doros showed how tools from statistical physics could predict how molecules arrange themselves and navigate the nanoscale pores within zeolites (Figure 16). In the process, they cultivated students who would put their own mark on this new branch of catalysis research: June,¹⁵⁸⁻¹⁶² Snurr,^{161,163-166} Maginn,¹⁶⁷⁻¹⁶⁹ and Coppens.¹⁷⁰⁻¹⁷²

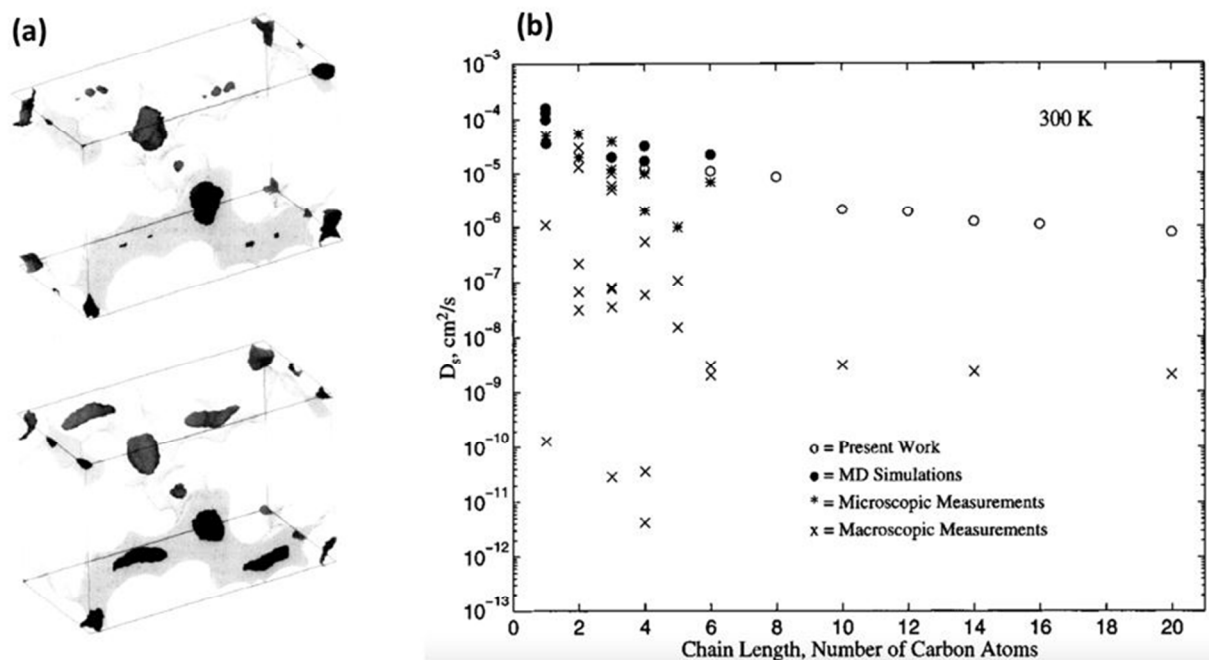


Figure 16. (a) Location of benzene molecules adsorbed in the pore space of silicalite-1 at 70°C, illustrating clearly defined sites in the pore space (dark regions: 95% probability), but also differences at low (top) and high loading (bottom).¹⁶⁵ (b) Self-diffusivity of n-alkanes as a function of chain length, at 300 K, comparing molecular dynamics and hierarchical simulations with experiments. The simulation strategy utilizes concepts from Brownian motion theory and transition state theory.¹⁶⁹

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3 The overarching finding from this work is that adsorption and transport within zeolites is
4 influenced by subtle heterogeneities in structure and composition and by the correlations
5 between adsorbate molecules at high loadings. These effects are not well captured by mean-field
6 models. Elsewhere in the catalysis world, van Santen, Sauer, Catlow and others were also
7 finding that Langmuir-Hinshelwood expressions and other mean-field relationships do not
8 adequately account for the local environments and interactions between adsorbed species on
9 surfaces. Especially at higher loadings, these local environments differ from the averages, and
10 may not be representative of the conditions relevant to heterogeneous catalysis. In the most
11 extreme cases, as poignantly illustrated by Ertl, deviations from mean field behavior give rise to
12 multiple steady states, transient behavior, and nonlinear waves.
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27 A third stage of Alex' theoretical work began in the mid-1990s, as *ab initio*
28 computational chemistry was growing in power, accuracy, and accessibility. Alex and his
29 colleague Arup Chakraborty saw an opportunity for catalysis research in new hybrid density
30 functionals and Pople basis sets. They were among the pioneers in using computational
31 chemistry, particularly density functional theory, to study adsorption and reactivity in zeolites.
32 Their work illustrated how these tools could be used to predict binding enthalpies, reaction
33 energies, activation barriers, and to make *ab initio* spectroscopic assignments.¹⁷³⁻¹⁷⁷
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44 Trout *et al.* used density functional theory to study the speciation of Cu^+ , $[\text{CuO}]^+$, and
45 $[\text{CuOH}]^+$ ions¹⁷³ and the thermochemistry of NO decomposition¹⁷⁴ in a Cu-ZSM-5 catalyst. Rice
46 *et al.* used DFT to understand the role of water in the demetallation of Cu-ZSM-5 and Co-ZSM-
47 5.¹⁷⁷ In their study of N_2O decomposition by Fe-ZSM-5 and Co-ZSM-5, Ryder *et al.* showed
48 how *ab initio* calculations of rate constants and equilibrium constants can facilitate the
49 construction of microkinetic models and the interpretation of experimental activation barriers.¹⁷⁸
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3 In these early computational studies, the number of intermediates and elementary steps was
4 limited by the difficulty of the electronic structure calculations and transition state optimizations.
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8 Alex' early computational studies, using tools from both molecular simulation and
9 quantum chemistry, gave him a unique perspective on the challenges ahead. His 2004 review
10 article, "Challenges for the application of quantum chemical calculations to problems in
11 catalysis",¹⁷⁹ identified four main challenges for the future. This review article marks the
12 beginning of a fourth stage in Alex' theoretical/computational work. The method development
13 challenges identified in Alex' article became a to-do list, not just for Alex, but also for the
14 computational catalysis efforts of many others. His method development goals and contributions
15 toward achieving them include:
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27 *(1) Accurate rate expressions for the elementary steps in catalysis.*
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29 Alex has shown that recrossing corrections to transition state theory are usually negligible
30 in catalysis,¹⁸⁰ but tunnelling¹⁸¹ and curve-crossing (non-adiabatic) rates can be
31 important.¹³⁶ He has also shown that non-equilibrium dynamics determine product
32 selectivity in acid (zeolite) catalysed cracking reactions.^{182,183}
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39 *(2) Accurate models of active sites and adsorbate interactions in zeolites.*
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41 Alex' early work used constrained cluster models for zeolites,^{175,177,184} but his more
42 recent work used larger models with state-of-the-art quantum chemistry and importance
43 sampling methods. Their efforts resulted in benchmark studies of hydrocarbon
44 adsorption that include electrostatics and dispersion through QM/MM embedding
45 schemes,¹⁸⁵ and re-optimized classical force fields for modelling alkanes in zeolites.^{186,187}
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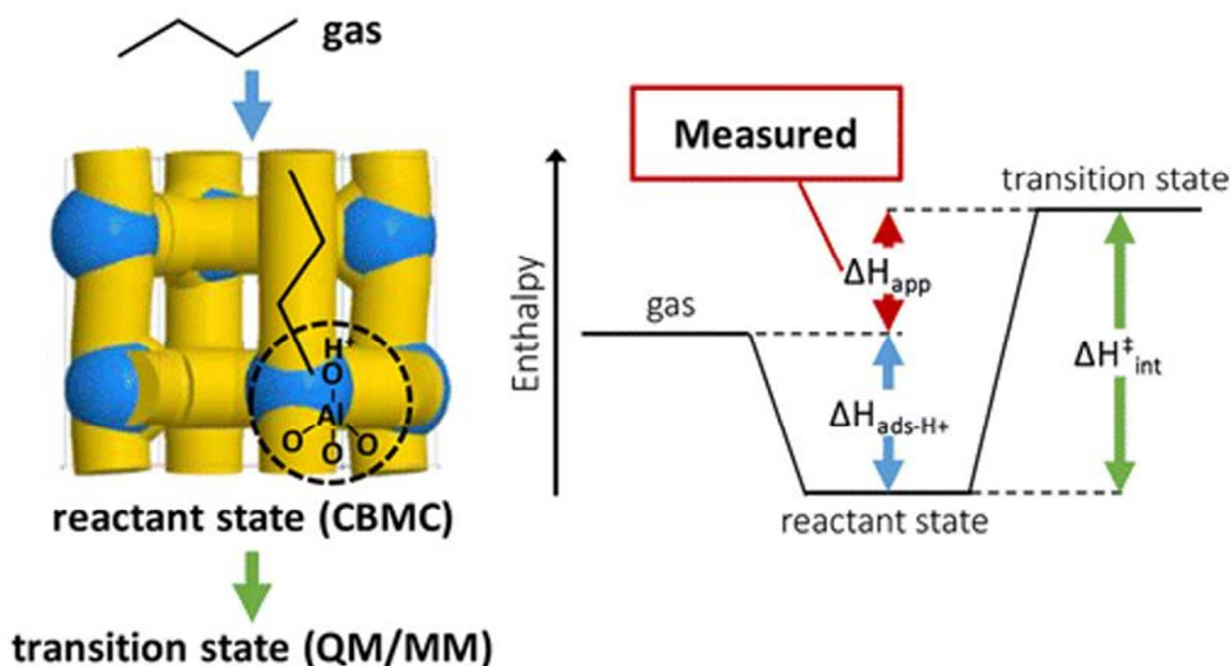
53 *(3) Efficient, automated methods for discovering reaction pathways and intermediates.* In
54 collaboration with Arup Chakraborty and Frerich Keil, Alex developed new transition
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3 state search algorithms with improved efficiency,¹⁸⁸⁻¹⁹⁴ improved fault tolerance,¹⁹⁵
4 biases to avoid rediscovery,¹⁹⁶ and automated path generation capabilities¹⁹⁷ to reduce
5 reliance on human effort and intuition. Some of these tools are now state-of-the-art
6 algorithms for discovering transition states and intermediates in complex reaction
7 networks.
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15 The above methodological advances from Alex' own research group and also from others
16 enabled studies of vastly more complex catalytic reaction networks and processes. In a series of
17 computational papers published between 2005 and 2008, Alex revisited his earlier studies of N₂O
18 and NO decomposition by Fe-ZSM-5. The works by Heyden *et al.*^{198,199} and Hansen *et al.*²⁰⁰
19 expanded the NO_x reduction mechanisms to include hundreds of elementary reactions at mono-
20 and bi-nuclear Fe sites. Their *ab initio* calculations and kinetic models explain the effects of
21 temperature, humidity, oxygen, and iron loading on the NO_x reduction activity of Fe-ZSM-
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34 An elegant series of computational studies from Alex' group examined hydrocarbon
35 cracking by acid zeolites. The overall cracking mechanism involves three steps: adsorption of
36 alkanes into the zeolite framework, association with an acid site, and the catalytic protonolysis
37 steps. Catalytic cracking, as Alex surely anticipated, was ideal for computational analysis:
38 industrially important, simple enough to permit comprehensive calculations, and sufficiently
39 complex to present unresolved questions. At the time of their computational studies, the overall
40 activation energy was known to depend on chain length primarily because of the adsorption step.
41 Alex, working with Berend Smit and Martin Head-Gordon, extended our understanding to the
42 more subtle effects of entropy, zeolite structure, and reaction dynamics.^{182,202,203} The analysis by
43 Swisher *et al.* is a blueprint for rigorous and thorough computational work.²⁰⁴ They used
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4 configurational bias Monte Carlo simulations to compute the adsorption enthalpy and entropy at
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6 reaction conditions for various alkanes, in various sites, and in several different zeolites. They
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8 combined the computed adsorption enthalpies and entropies with overall activation parameters
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10 from experiment to obtain activation enthalpy and entropy estimates for the catalytic steps (see
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12 Figure 17). Swisher et al. also used DFT to predict rates for the chemical step.²⁰⁴ Computed and
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14 measured overall rates (for the FAU zeolite) agreed within a factor of about 100 from propane to
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16 n-hexane. Later calculations by Sharada *et al.*²⁰⁵ and Janda *et al.*²⁰⁴ improved the calculations
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18 with better DFT calculations and better zeolite models.
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Figure 17. Combined adsorption enthalpies and entropies from Monte Carlo simulations (blue) with overall activation parameters from experiment (red) to infer activation parameters for the chemical step (green). An independent confirmation of the overall cracking mechanism can then be obtained by using DFT calculations for the elementary cracking step(s).²⁰²

Alex in Practice

This methodical approach reveals the long-term, systematic thinking of Alex Bell as a scientist. Patient development of new computational and experimental tools, applying them step

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3 by step to pertinent problems in structured catalysts, allow him to, over time, construct a more
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5 and more complete picture of the problem at hand, solving outstanding questions that lead from
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7 science, to development, and to design, as outlined in Reference 75.
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10 One thing that stands out when reading Alex Bell's published work is his exceptional
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12 writing skill. Both in original journal articles and in perspective articles and reviews, his writing
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14 is clear and concise, with well-chosen words and easy sentences. The presentation of the
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16 scientific story is well crafted from Abstract through to Conclusions. The main findings, as well
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18 as the motivation for the work jump right off the page, supported by clear scientific arguments,
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20 and illustrated by the perfect number of tables and figures. Alex devoted much of his time
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22 teaching scientific writing to his graduate students. All of them benefited greatly from his
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24 mentorship, and throughout their careers employed to advantage the writing and oral skills
25
26 learned from Professor Bell.
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31 As a former postdoctoral researcher during the time that Bell was Dean of the College of
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33 Chemistry at UC Berkeley, one of the authors (Marc-Olivier Coppens) was struck by the way in
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35 which work for the College and research time would be perfectly separated, in space (different
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37 offices) and time (regular appointments). In this, he was as systematic as the way in which he
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39 would conduct his research. For example, on any Wednesday, Alex Bell would have research
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41 appointments in his Departmental office during the morning (Marc-Olivier's were at 10am), then
42
43 move to the Dean's Office for the afternoon, and typically make a round in the evening, where
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45 he would sometimes drop into the office for an extra conversation. Alex Bell was always poised,
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47 well organised and prepared; he listened carefully and asked excellent questions, leading to
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49 effective discussions. This made a great impact on Marc-Olivier for organizing his work later on
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3 in his career, especially in combining his current positions as Head of Department, Centre
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5 Director, scientific research and time for teaching and students.
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8 Those who haven't worked with Alex may wonder how anyone can have such
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10 tremendous breadth, but anyone who has written a paper with him has seen the recipe. When
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12 Alex sits down with collaborators or students to revise a manuscript, he isn't just there to finish
13
14 the paper. He wants to assimilate and master the full skillset of his co-author. In the familiar
15
16 parts of a manuscript, he asks razor sharp questions about wording and internal consistency. But
17
18 when he encounters a part of the manuscript with ideas, principles, or methods that are new, he
19
20 asks different types of questions. He wants to know in full detail how it works and how it relates
21
22 to what he already knows. He is a fast study. After one long meeting, Alex can not only use the
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24 new ideas but he can give an impromptu lesson to the next student.
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29 As a PhD advisor, Alex is a fountain of refreshing, child-like enthusiasm. He typically
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31 arrives a few minutes late to meetings with his PhD students (we called it Berkeley time). He
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33 hustles in with an excited grin, wringing his hands as though he was next up to bat, and then
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35 dives right into the technical discussion. His excitement over new results was a wonderful
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37 source of motivation.
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42 **Summary**

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44 We have enjoyed putting together even this modest number of vignettes to help celebrate
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46 Alex's contributions to catalysis and reaction engineering – both as a technical field and as a
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48 scholarly community. At the occasion of his 70th birthday celebration in Berkeley Alex
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50 commented that he was drawn to Berkeley because John Prausnitz told him on a visit that 'here
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52 students do not work for us, we collaborate with them'. He did just that – collaborate with his
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54 students and postdoctoral fellows.
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Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI:xxxxxxxxxxxxx.

A list of Alex Bell's publications (PDF).

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List of Figures

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- Figure 1. Sequence of elementary reactions expected to occur when a hydrocarbon monomer enters a discharge zone of a DC plasma reactor.⁸
- Figure 2. The rate of polymerization, which is the sum of gas and surface propagation steps was determined by solving the material balance equations that describe the monomer and free radical concentrations as a function of axial and radial position in the flow reactor. Predicted and experimental rates of polymer deposition as a function of monomer flow rate are one illustration of this model fit.⁸
- Figure 3. Dependence of the Rh-NCO and Rh-NO^{δ-} absorbance bands as a function of NO partial pressure. During the reduction of NO by CO over a Rh/SiO₂ catalyst an isocyanate group is formed on the surface of the rhodium microcrystallites. The dependence of the Rh-NCO band intensity on partial pressures of NO and CO can be interpreted with the aid of the reaction mechanism.²⁴
- Figure 4. Transient response and isotope tracer studies revealed two distinctly different forms of carbon formed on Ru/SiO₂ during CO hydrogenation. C_α is highly reactive and is the principle precursor to C₂₊ hydrocarbons and methane. C_β accumulates slower than C_α and resides on Ru and the support.²⁷
- Figure 5. Proposed mechanism of hydrocarbon synthesis from CO and H₂.¹⁸
- Figure 6. Reaction scheme for the rapid equilibration of CO into adsorbed CO and the interconversion of dissociated CO into the various forms of carbon.⁶¹
- Figure 7. Arrhenius plots for the formation of methanol at 10 atm and a H₂/CO ratio of 3.⁶⁹
- Figure 8. Schematic illustration of the interaction of the Pd with the support.⁴⁴
- Figure 9. Illustration of clathrate organization around a template cation.⁹⁸

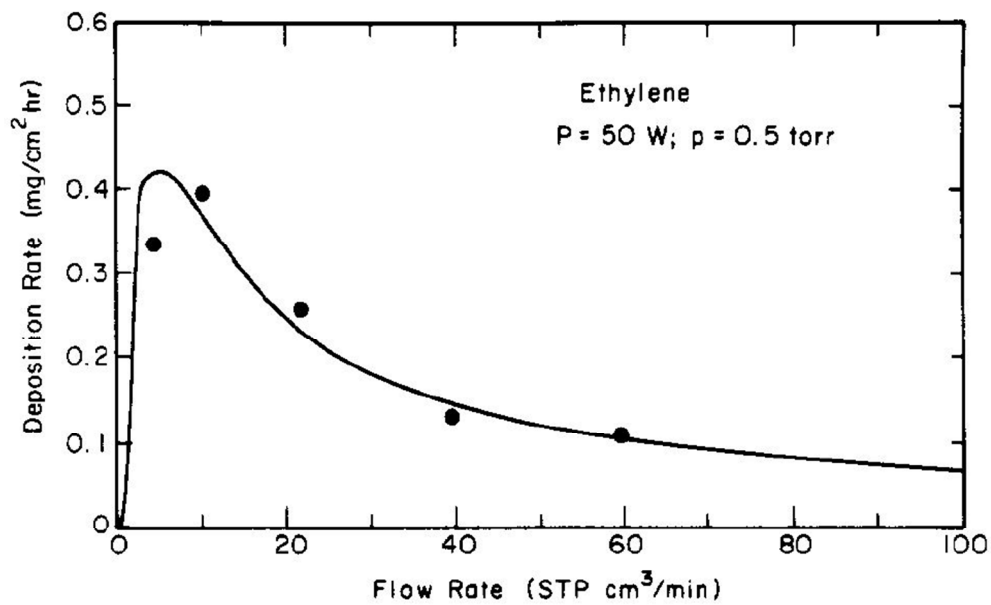
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3 Figure 10. Assembly of clathrates into a prospective nucleus.⁹⁸
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6 Figure 11. Quantitative ²⁹Si MAS NMR patterns, offering key evidence pointing toward a
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8 comprehensive description of the silicalite synthesis induction period.¹⁰²
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10 Figure 12. Mechanistic scheme for the TPD experiment on 3.4V/MCM-48.¹³⁰
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12 Figure 13. Mechanistic scheme for the TPO experiment on 3.4V/MCM-48.¹³⁰
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14 Figure 14. Model of the VO_x/SiO₂ catalytic site.¹³¹
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17 Figure 15. Model of the VO_x/TiO₂ catalytic site. The location of the O-vacancy is circled.¹³⁵
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20 Figure 16. (a) Location of benzene molecules adsorbed in the pore space of silicalite-1 at 70°C,
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22 illustrating clearly defined sites in the pore space (dark regions: 95% probability),
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24 but also differences at low (top) and high loading (bottom).¹⁶⁵ (b) Self-diffusivity of
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26 n-alkanes as a function of length, at 300 K, comparing hierarchical simulations with
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28 experiments. The simulation strategy utilizes concepts from Brownian motion theory
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30 and transition state theory.¹⁶
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34 Figure 17. Combined adsorption enthalpies and entropies from Monte Carlo simulations (blue)
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36 with overall activation parameters from experiment (red) to infer activation
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38 parameters for the chemical step (green). An independent confirmation of the
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40 overall cracking mechanism can then be obtained by using DFT calculations for the
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42 elementary cracking step(s).²⁰²
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Table I
Reaction Mechanism for Plasma Polymerization of Unsaturated Hydrocarbons^a

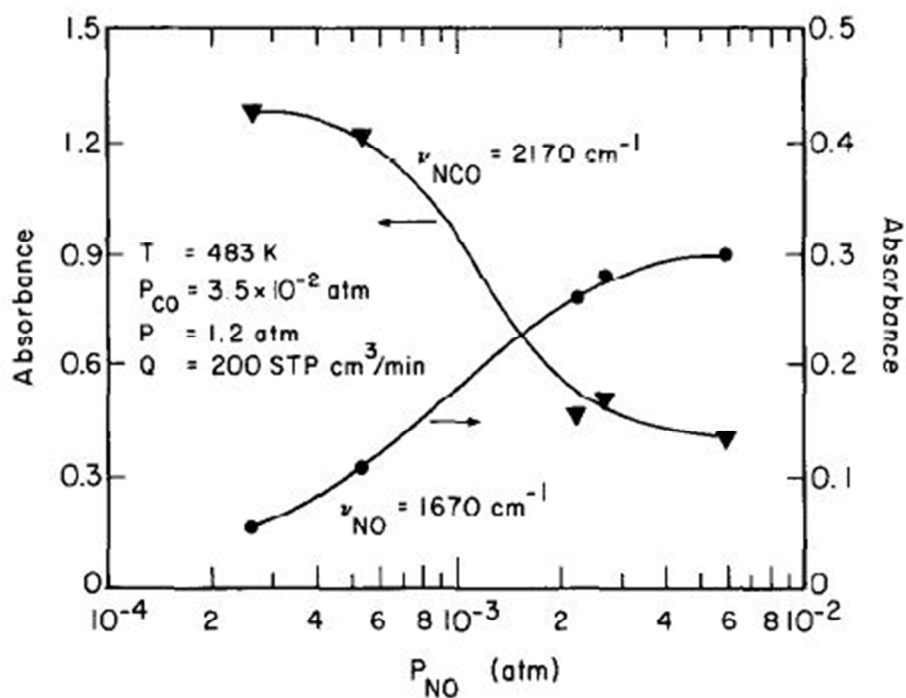
Initiation	Adsorption	Termination
1. $e + M_g \rightarrow M'_g + H_2 + e$		13. $R'_{gn} + H \cdot \rightarrow P_{gn}$
2. $e + M_g \rightarrow M'_g + 2H \cdot + e$	7. $S + \begin{Bmatrix} M_g \\ M'_g \end{Bmatrix} \rightarrow \begin{Bmatrix} M_s \\ M'_s \end{Bmatrix}$	14. $R'_{gm} + R'_{gn} \rightarrow P_{gm+n}$
3. $e + M_g \rightarrow 2R'_{gn} + e$	8. $S + H \cdot \rightarrow H_s$	15. $R'_{gm} + R'_{sn} \rightarrow P_{sm+n}$
4. $e + H_2 \rightarrow 2H \cdot + e$	9. $S + R'_{gn} \rightarrow R'_{sn}$	16. $R'_{sm} + R'_{sn} \rightarrow P_{sm+n}$
Propagation (homogeneous)	Propagation (heterogeneous)	Reinitiation
5. $H \cdot + \begin{Bmatrix} M_g \\ M'_g \end{Bmatrix} \rightarrow R'_{g1}$	10. $R'_{sn} + \begin{Bmatrix} M_g \\ M'_g \end{Bmatrix} \rightarrow R'_{sn+1}$	17. $e + P_{gm+n} \rightarrow R'_{gm} + R'_{gn}$
6. $R'_{gn} + \begin{Bmatrix} M_g \\ M'_g \end{Bmatrix} \rightarrow R'_{gn+1}$	11. $R'_{sn} + \begin{Bmatrix} M_s \\ M'_s \end{Bmatrix} \rightarrow R'_{sn+1}$	18. $P_s \xrightarrow{e, h\nu, I^+} R'_{sn} + R'_{sn}$
	12. $H_s + H \cdot \rightarrow H_2$	19. $H \cdot + P_{gn} \rightarrow R'_{gn} + H_2$
		20. $H \cdot + P_{sn} \rightarrow R'_{sn} + H_2$

^a M_g , primary monomer; R'_{g} , gas phase free radical; P_g , gas phase oligomer; M'_g , derivative monomer; R'_{s} , surface free radical; P_s , polymer fragment; H , hydrogen atom.

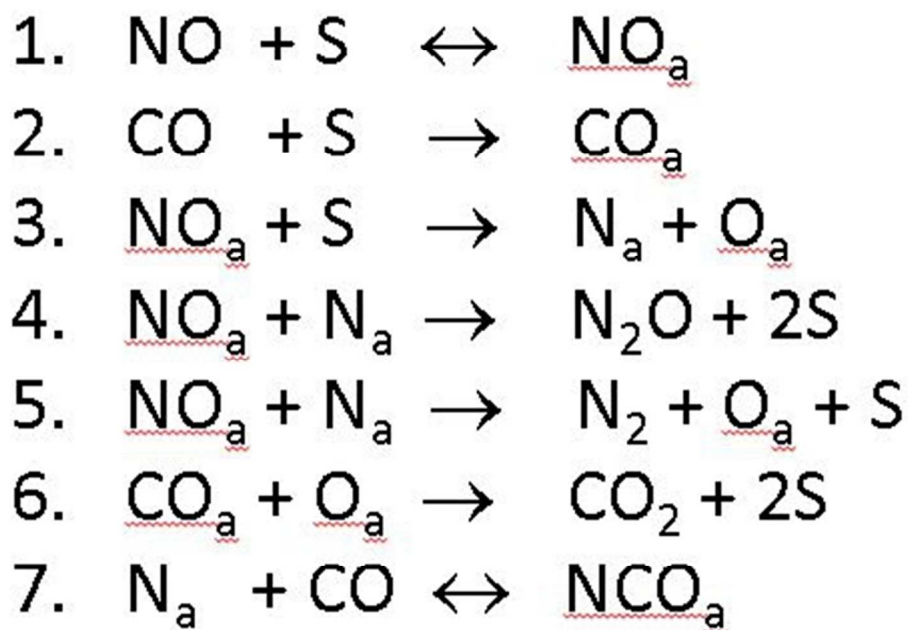
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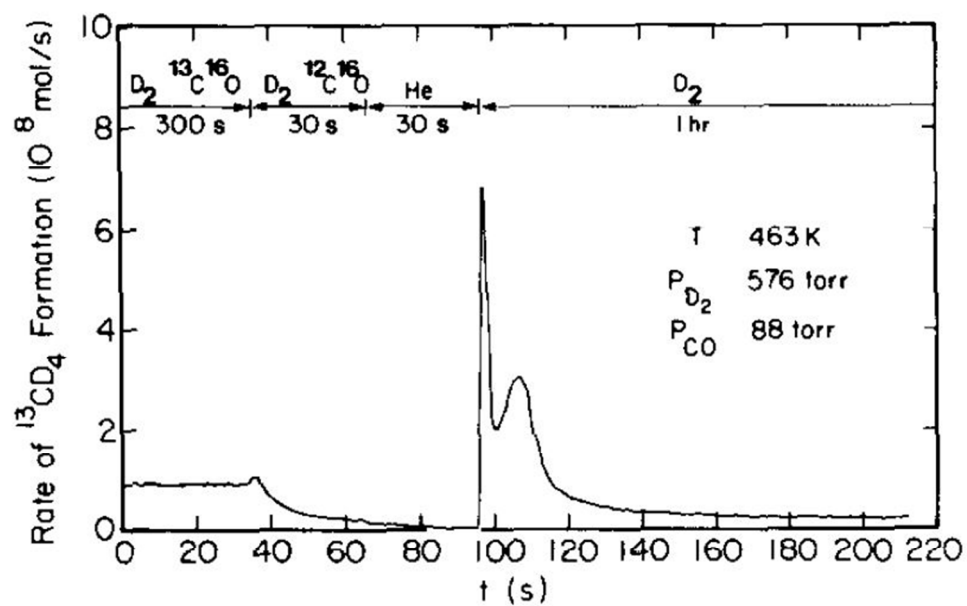
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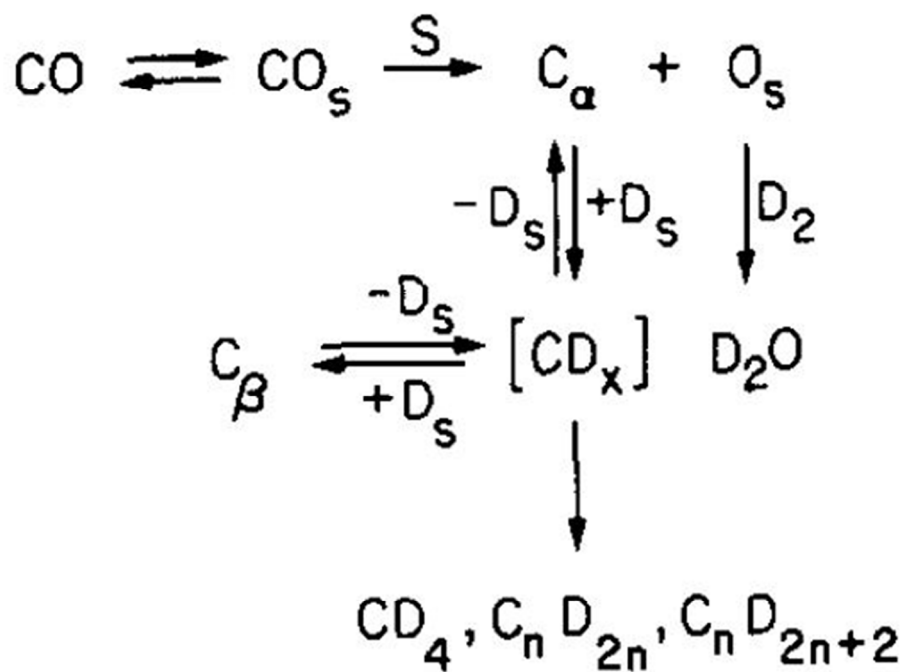
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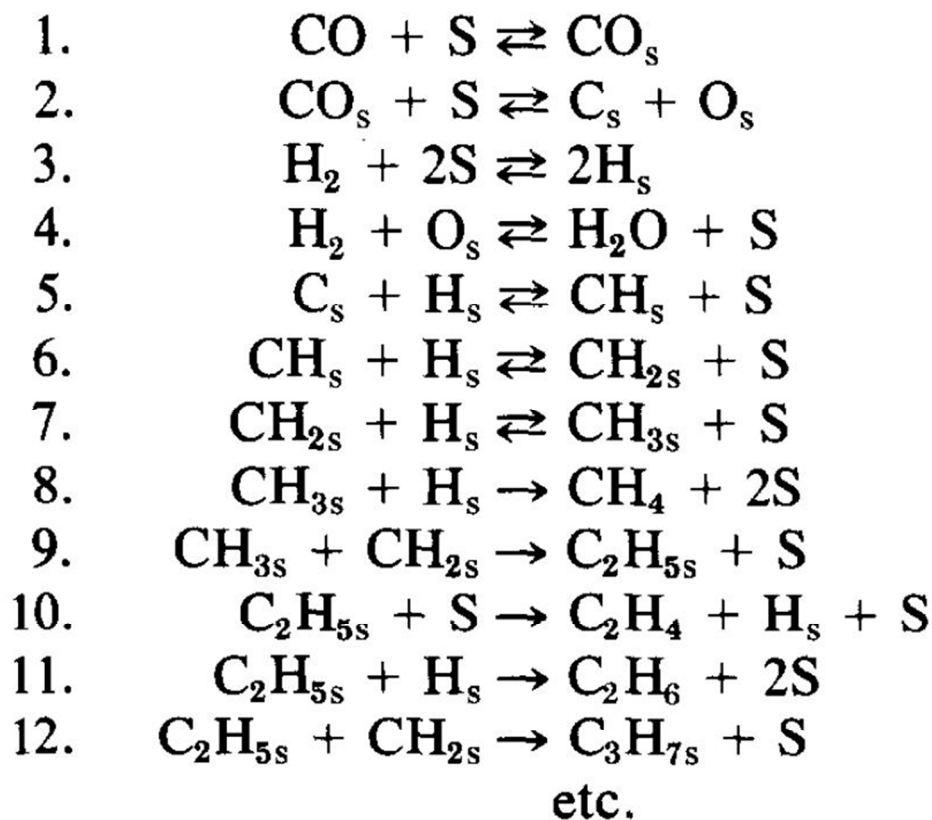
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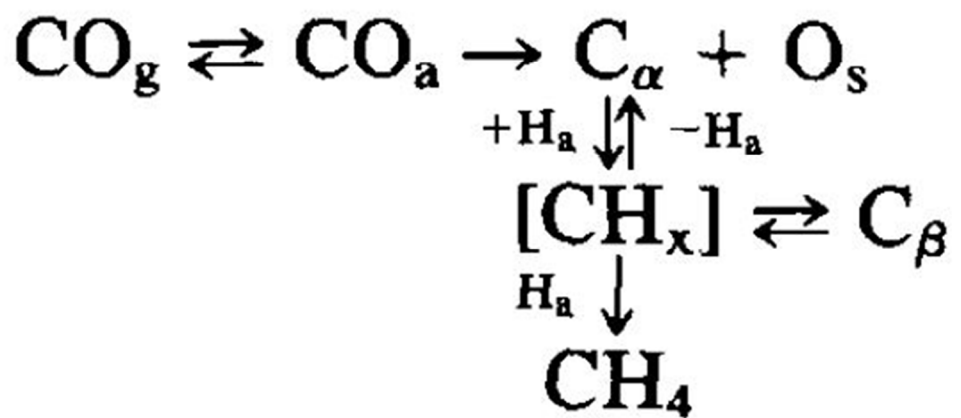
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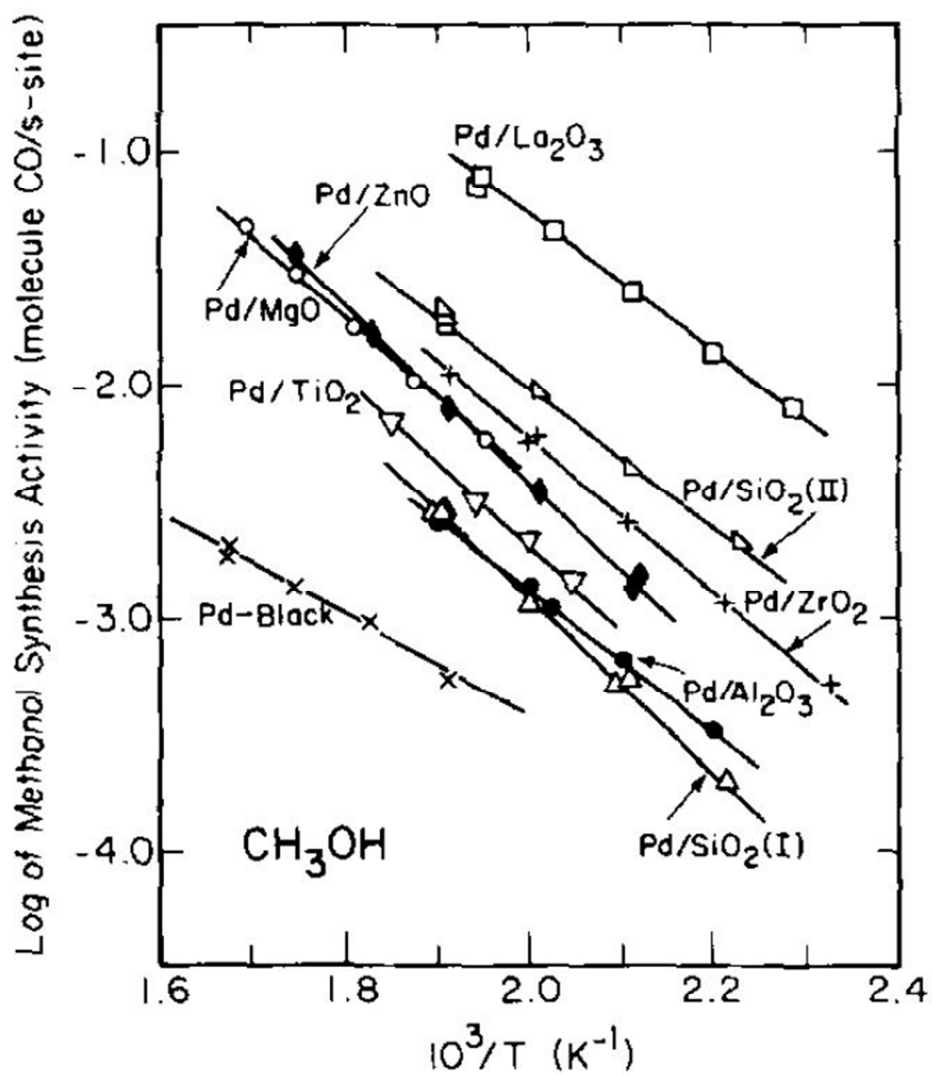
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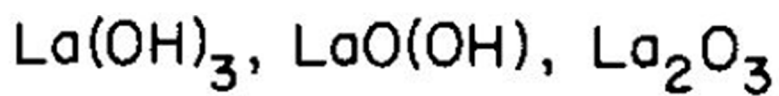
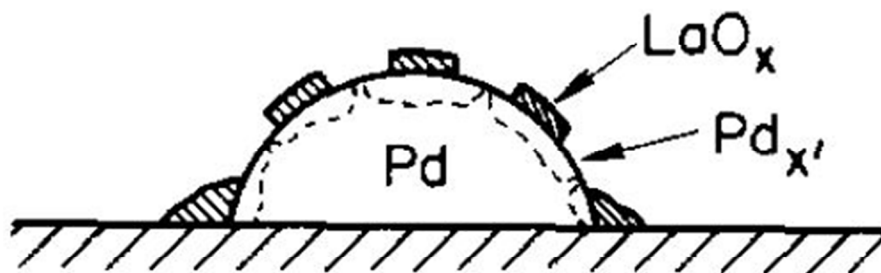
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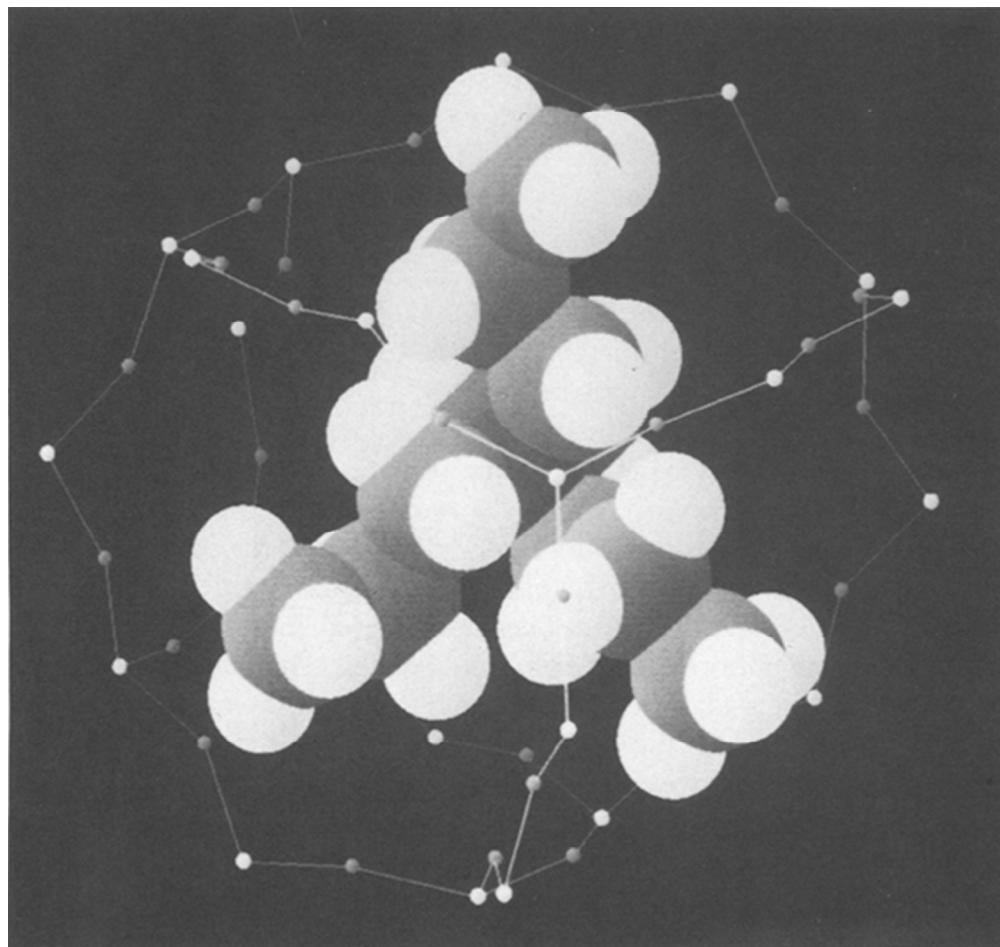
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191x211mm (96 x 96 DPI)

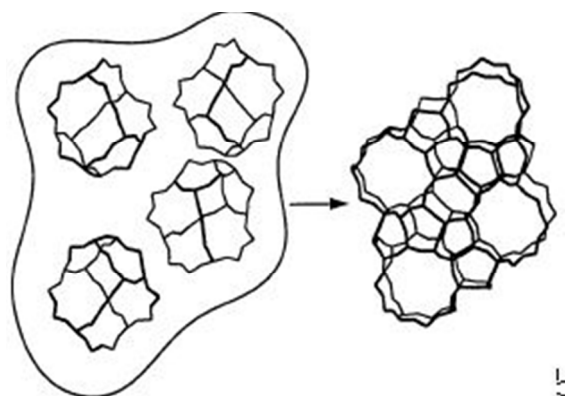


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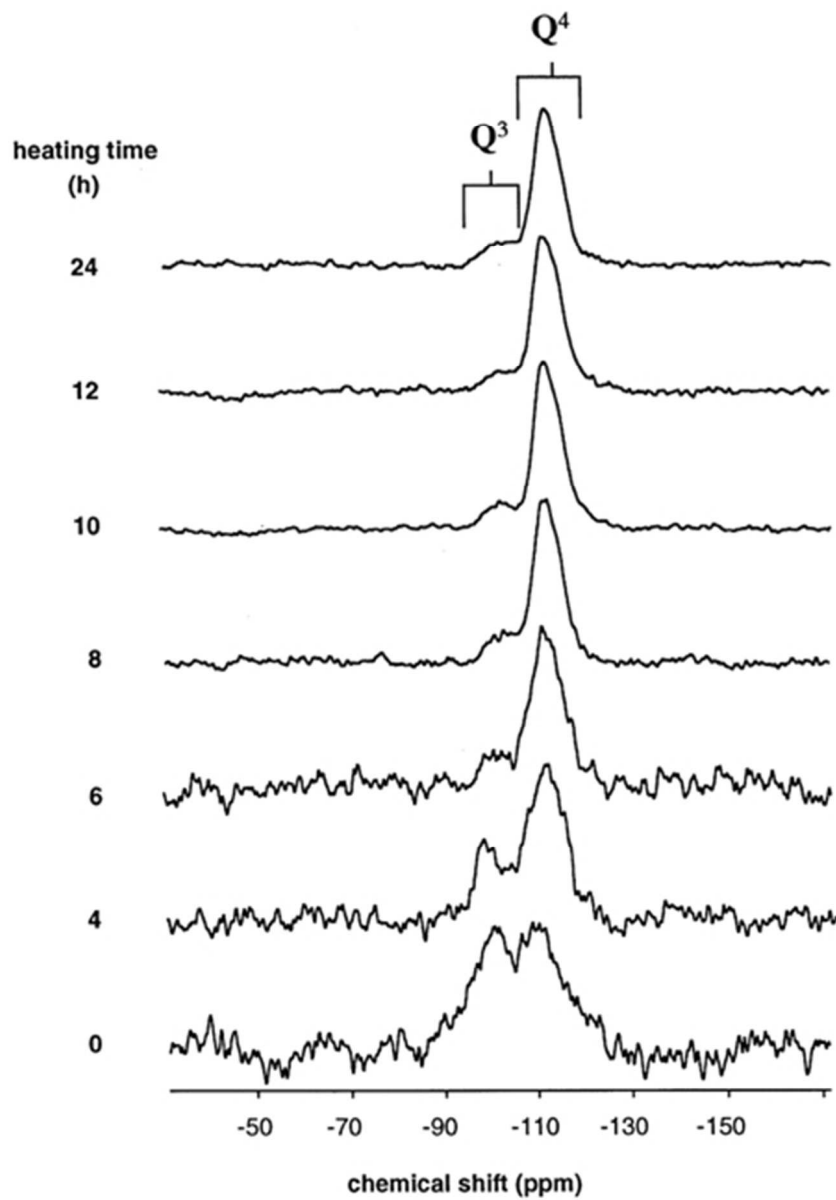


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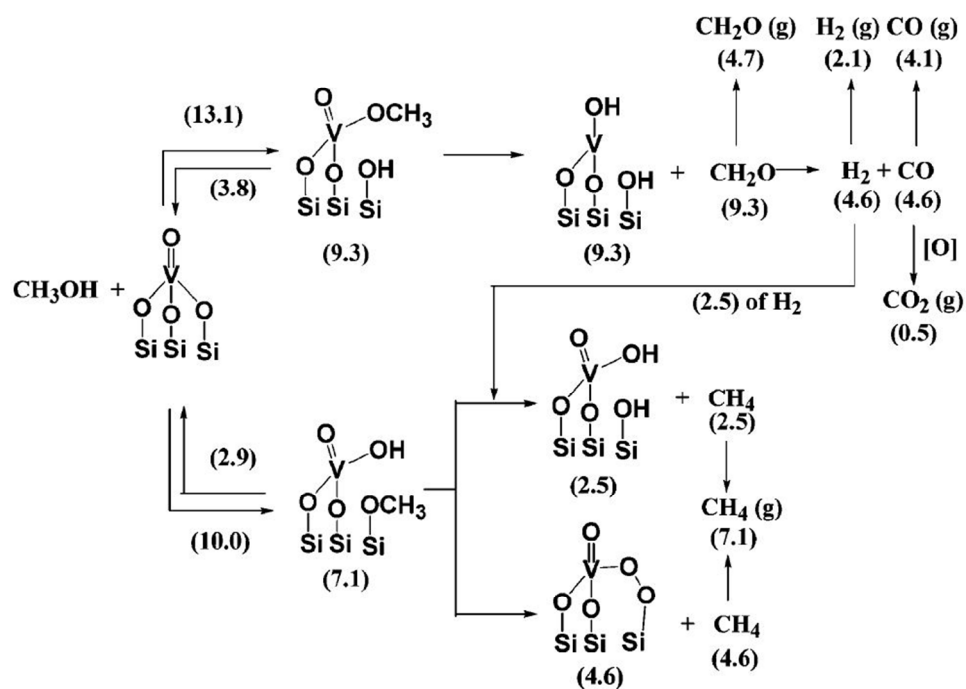
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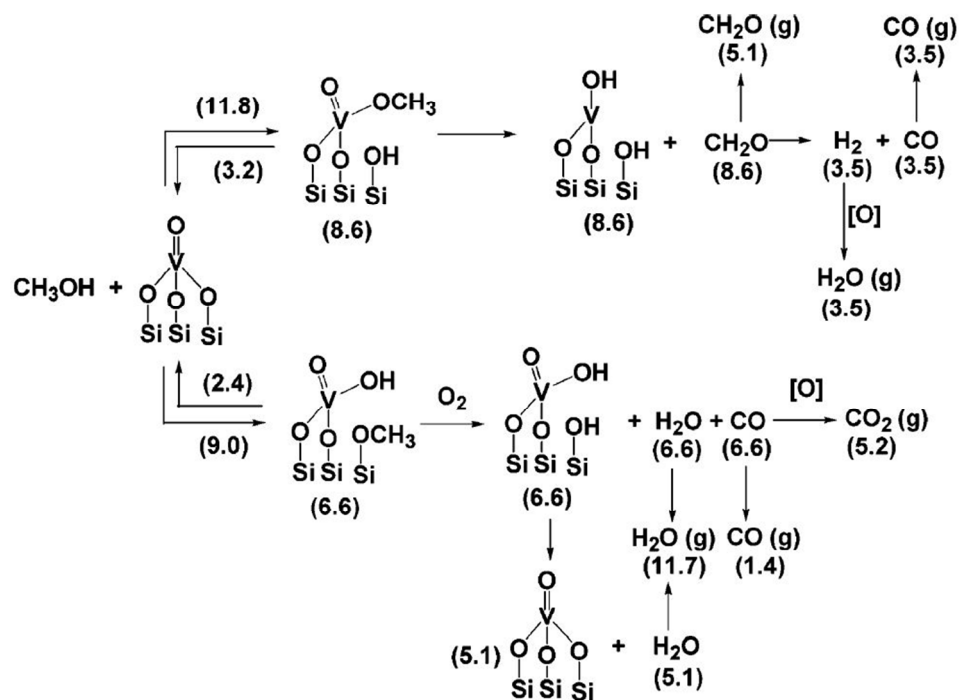
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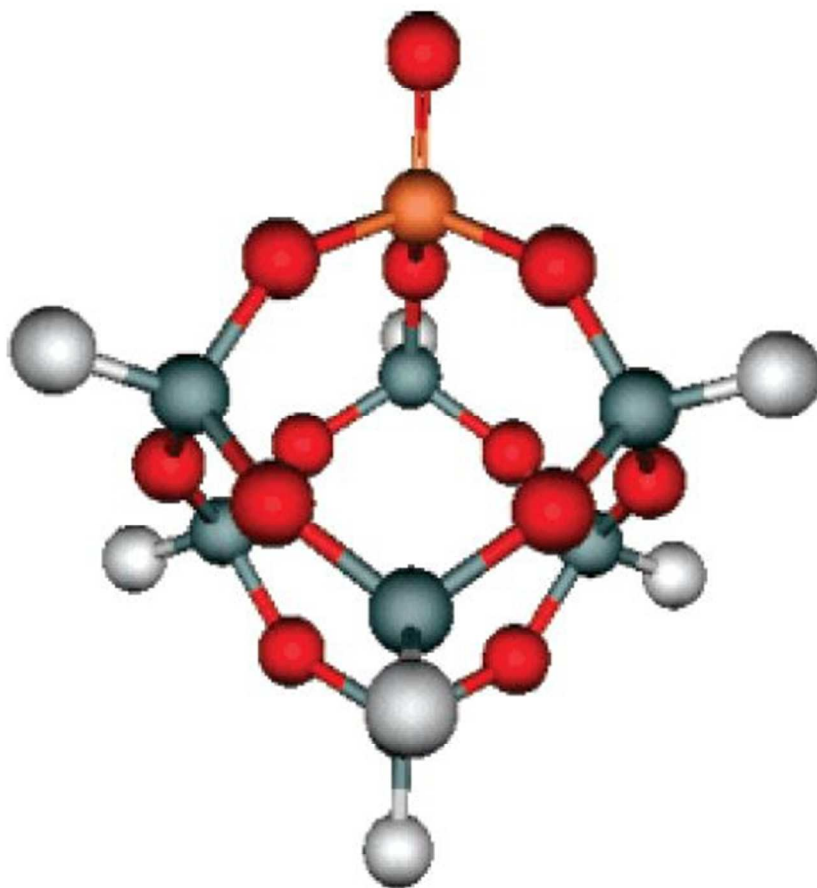


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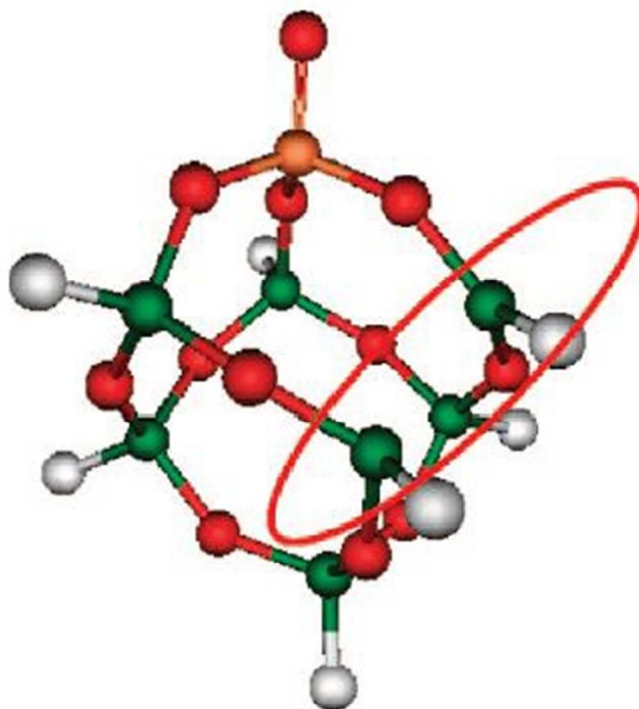
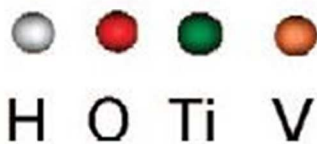
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H O Si V

153x191mm (96 x 96 DPI)

**1d**

97x130mm (96 x 96 DPI)

